Supporting Information

1. Materials and instrumentation

Chemicals were commercially available and used as received. Analytical thin-layer chromatography (TLC) was carried out on silica gel 60 F254 (Merck) and visualized under UV irradiation (a wavelength of 254 nm). Column chromatography was performed on silica gel (230–400 mesh or 37–63 μm). Analyses of gas chromatography (GC) were obtained from a Shimadzu GC 2010-Plus equipped with a flame ionization detector (FID) and an SPB-5 column. Runs of gas chromatography - mass spectrometry (GC-MS) were carried out on a Shimadzu GCMS-QP2010Ultra with a ZB-5MS column. The results of NMR were recorded on Bruker AV 500 and 600 spectrometers using the residual solvent peak as a reference. Chemical shifts were provided in parts per million (ppm). The abbreviations used to explain multiplicities are as follows: s = singlet, d = doublet, t = triplet, q = quartet, dd = doublet of doublets, bs = broad singlet, and m = multiplet. Coupling constants were reported in Hertz (Hz). High resolution mass spectrometry (HR-MS) spectra were recorded on an Agilent HPLC 1200 Series coupled to a Bruker micrOTOF-QII. Mass spectrometry was performed in the positive electrospray ionization (ESI+) mode.

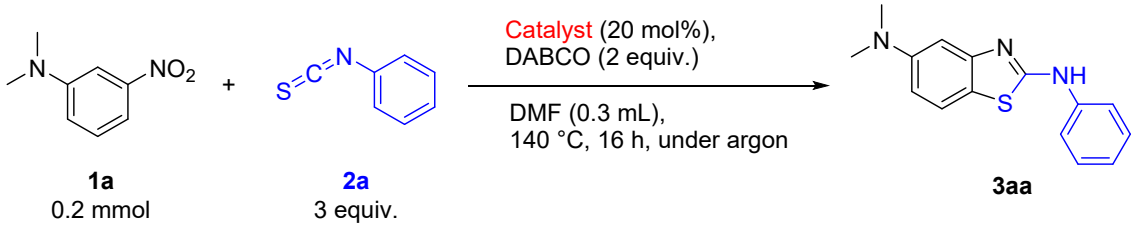
2. Optimization of reaction conditions

2.1. General procedure for screening process

A mixture of a *N*,*N*-dimethyl-3-nitroaniline (**1a**), phenyl isothiocyanate (**2a**), catalyst, base and solvent (if necessary) was added to a dried 4 mL vial equipped with a magnetic stir bar. The mixture was placed into a bath preheated to 140 °C and stirred for 16 h under argon. After the reaction was completed, the mixture was left to cool to room temperature. Diphenyl ether was then added to the resulting mixture as an internal standard. An aliquot of the resulting mixture was added to a test tube containing brine (2 mL) then extracted with ethyl acetate (3 x 2 mL). The combined organic components were dried over anhydrous Na_2SO_4 and filtered. The resulting solution was analyzed by GC to determine the GC yield of product **3aa** using diphenyl ether as internal standard.

2.2. Screening reaction conditions

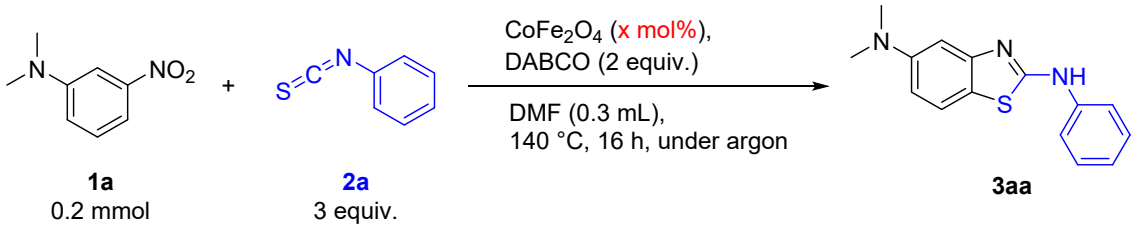
2.2.1. Catalysts



Entry	catalyst (20 mol%)	yield of **3aa** (%)[a]
1	CuI	6
2	$Cu(OAc)_2$	16
3	$FeSO_4.7H_2O$	39
4	$FeCl_3$	29
5	Fe_3O_4	66
6	**$CoFe_2O_4$**	**76**

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), catalyst (20 mol%), DABCO (0.4 mmol), DMF (0.3 mL), 16 h, 140 °C, under argon. Yields are GC yields.

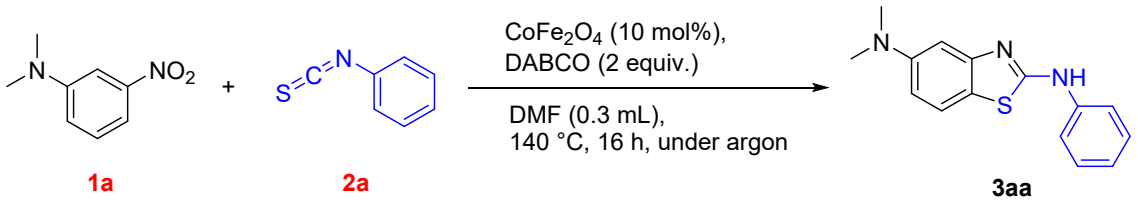
2.2.2. Catalyst loading



Entry	$CoFe_2O_4$ (mol%)	yield of **3aa** (%)[a]
1	0	15
2	5	58
3	**10**	**73**
4	20	76

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), $CoFe_2O_4$ (x mol%), DABCO (0.4 mmol), DMF (0.3 mL), 16 h, 140 °C, under argon. Yields are GC yields.

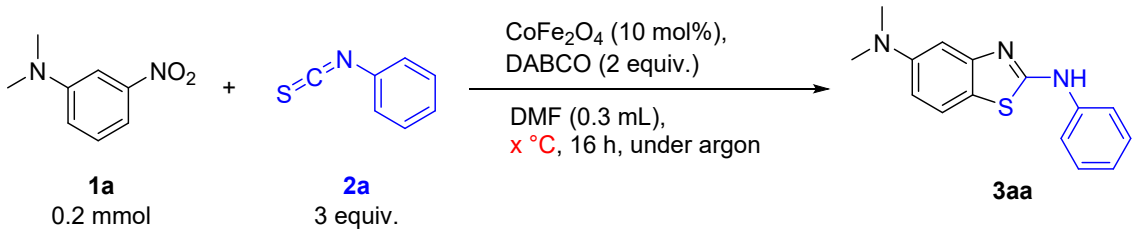
2.2.3. *Molar ratio of reactants*



Entry	**1a** : **2a**	yield of **3aa** (%)[a]
1	2 : 1	26
2	1 : 1	25
3	1 : 2	57
4	**1 : 3**	**73**
5	1 : 4	73

[a]Reaction conditions: limiting reactant (0.2 mmol), $CoFe_2O_4$ (10 mol%), DABCO (0.4 mmol), DMF (0.3 mL), 16 h, 140 °C, under argon. Yields are GC yields.

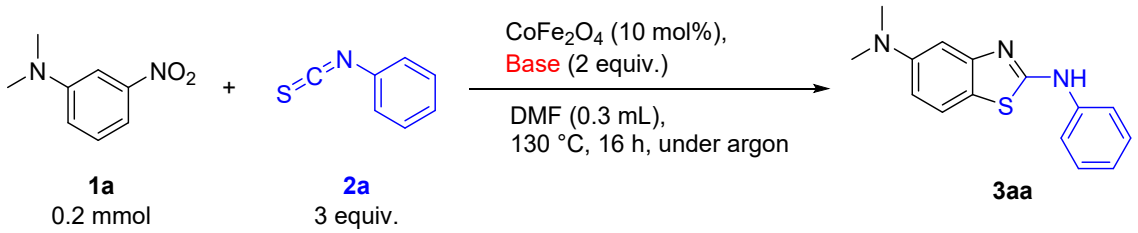
2.2.4. *Temperature*



Entry	temperature (°C)	yield of **3aa** (%)[a]
1	100	12
2	120	48
3	**130**	**75**
4	140	73

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), $CoFe_2O_4$ (10 mol%), DABCO (0.4 mmol), DMF (0.3 mL), 16 h, under argon. Yields are GC yields.

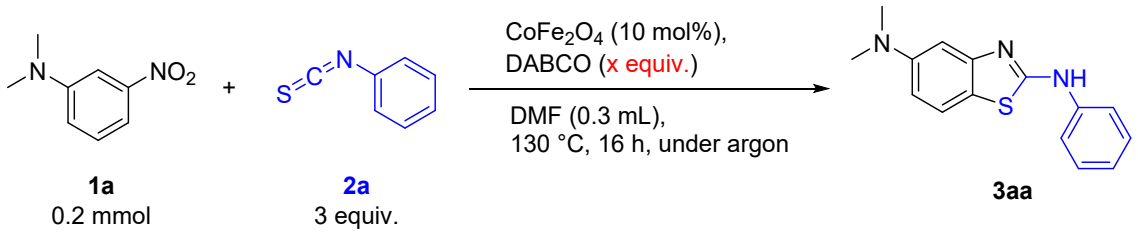
2.2.5. Bases



Entry	Base	yield of **3aa** (%)[a]
1	None	7
2	K_2CO_3	5
3	Cs_2CO_3	trace
4	NaOH	61
5	KOH	20
6	*t*-BuOK	5
7	NaOAc	8
8	4-DMAP	27
9	*N*,*N*′-dimethylpiperazine	64
10	DIPEA	41
11	**DABCO**	**75**
12	DBU	25
13	*N*-methylpiperidine	29
14	*N*-morpholine	15

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), $CoFe_2O_4$ (10 mol%), base (0.4 mmol), DMF (0.3 mL), 130 °C, 16 h, under argon. Yields are GC yields.

2.2.6. *Base amount*



1a
0.2 mmol

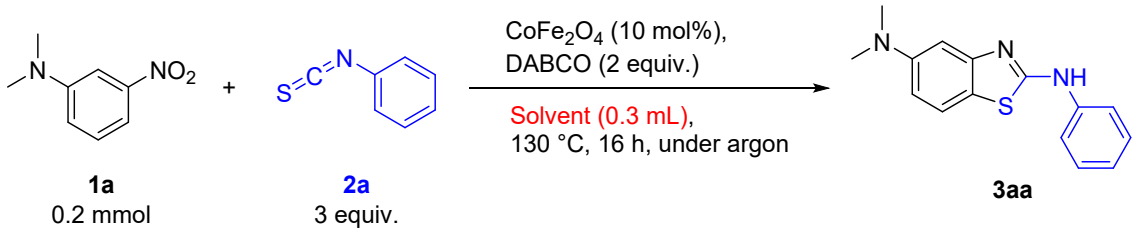
2a
3 equiv.

3aa

Entry	base amount (equiv.)	yield of **3aa** (%)[a]
1	0	7
2	1	63
3	**2**	**75**
4	3	75

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), $CoFe_2O_4$ (10 mol%), DABCO, DMF (0.3 mL), 130 °C, 16 h, under argon. Yields are GC yields.

2.2.7. *Solvents*



1a
0.2 mmol

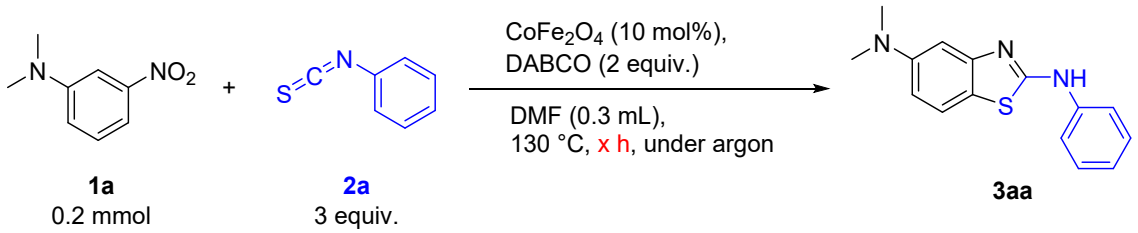
2a
3 equiv.

3aa

Entry	solvent	yield of **3aa** (%)[a]
1	DMSO	14
2	*N*-Methyl-2-pyrrolidone	58
3	**DMF**	**75**
4	chlorobenzene	44

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), $CoFe_2O_4$ (10 mol%), DABCO (0.4 mmol), solvent (0.3 mL), 130 °C, 16 h, under argon. Yields are GC yields.

2.2.8. Reaction time



1a
0.2 mmol

2a
3 equiv.

3aa

Entry	time (h)	yield of **3aa** (%)[a]
1	4	45
2	8	51
3	12	59
4	16	75
5	**24**	**80**
6	48	79

[a]Reaction conditions: *N*,*N*-dimethyl-3-nitroaniline (0.2 mmol), PhNCS (0.6 mmol), $CoFe_2O_4$ (10 mol%), DABCO (0.4 mmol), DMF (0.3 mL), 130 °C, under argon. Yields are GC yields.

3. Studying of reusability

The reaction was carried out at 130 °C under argon for 24 h, utilizing DABCO (2 equiv.) with DMF (0.3 mL), in the presence of 10 mol% catalyst $CoFe_2O_4$. After each run, the reaction vial was placed on a neodymium magnet for 15 min and the solution was removed. The residual solid was washed carefully with copious amounts of solvents including hexanes, ethyl acetate and ethanol. The ensuing solid was dried at 200 °C under vacuum for 12 h and reused in subsequent runs.

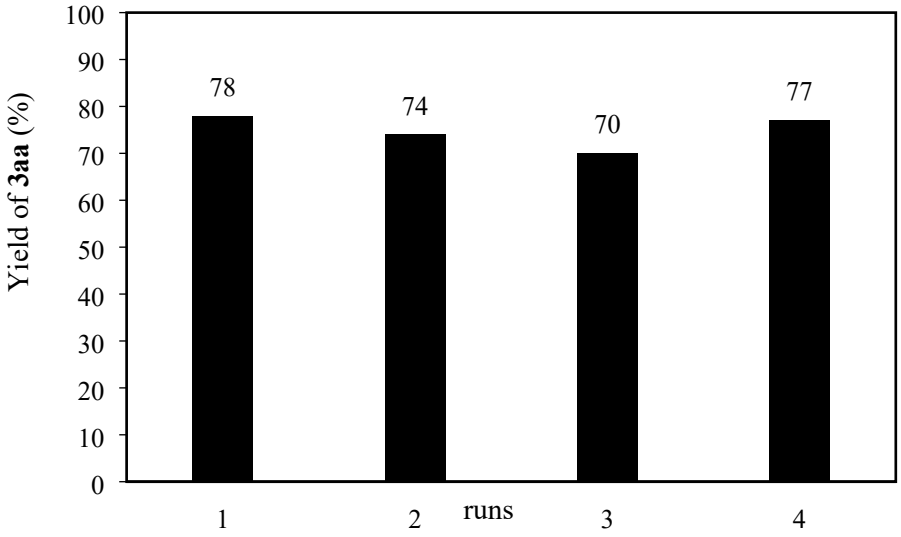


Figure S1. Reusability of the cobalt ferrite catalyst.

4. Synthesis of 2-aminobenzothiazoles

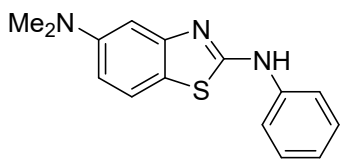
4.1. General procedure

A mixture of a *N*,*N*-dialkyl-3-nitroanilines derivative (0.2 mmol), an aryl isothiocyanate (0.6 mmol), DABCO (0.4 mmol, 44.8 mg), $CoFe_2O_4$ (10 mol%, 4.7 mg) and DMF (0.3 mL) was added to a dried 4 mL vial equipped with a magnetic stir bar. The mixture was placed into a bath preheated to 130 °C and stirred for 24 h under argon. The crude reaction mixture was diluted with ethyl acetate (30 mL), filtered and then washed with brine (4 × 10 mL). The combined organic phases were dried over anhydrous Na_2SO_4, filtered, and concentrated. Purification by column chromatography yielded the desired product.

For a 1 mmol run of **3aa**: to a dried 8 mL vial equipped with a magnetic stir bar were added *N*,*N*-dimethyl-3-nitroaniline **1a** (1 mmol, 166.9 mg), phenyl isothiocyanate **2a** (3 mmol, 410 mg), DABCO (2 mmol, 226.5 mg), and $CoFe_2O_4$ (0.1 mmol, 24 mg) and DMF (1 mL) . The vial was then placed in a bath preheated to 130 °C and stirred for 24 h under argon. The crude reaction mixture was diluted with ethyl acetate (80 mL), filtered and then washed with brine (4 × 30 mL). The combined organic phases were dried over anhydrous Na_2SO_4, filtered, and concentrated. Purification by column chromatography (toluene/dichloromethane 1:1) yielded **3aa** (226.9 mg, 84% yield) as a yellow solid.

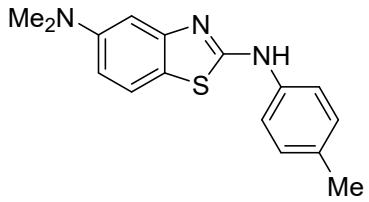
4.2. Characterization of all products

N^5,N^5-Dimethyl-N^2-phenylbenzo[*d*]thiazole-2,5-diamine (3aa)



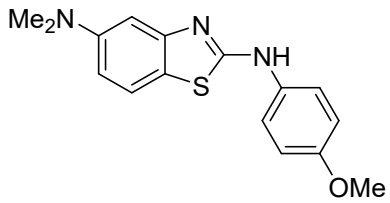
Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.4 mg), phenyl isothiocyanate **2a** (0.6 mmol, 84.2 mg), $CoFe_2O_4$ (0.02 mmol, 4.9 mg), DABCO (0.4 mmol, 45.3 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/dichloromethane 1:1), 46.9 mg (87% yield) of **3aa** was obtained as a yellow solid. R_f = 0.34 (hexanes/ethyl acetate 4:1), mp 159 – 161 °C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.10 (s, 1H), 7.51 (d, J = 7.7 Hz, 2H), 7.43 (d, J = 8.7 Hz, 1H), 7.39 (t, J = 7.9 Hz, 2H), 7.14 (t, J = 7.4 Hz, 1H), 6.95 (d, J = 2.1 Hz, 1H), 6.66 (dd, J = 8.7, 2.2 Hz, 1H), 2.93 (s, 6H). ^{13}C NMR (125 MHz, $CDCl_3$) δ 165.7, 152.9, 150.4, 140.3, 129.6, 124.2, 120.9, 120.5, 117.9, 109.6, 103.7, 41.3. HRMS (ESI) *m*/*z* calcd for $C_{15}H_{15}N_3S$ $[M+H]^+$: 270.1059, found: 270.1061.

N^5,N^5-Dimethyl-N^2-(*p*-tolyl)benzo[*d*]thiazole-2,5-diamine (3ab)



Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.3 mg), 1-isothiocyanato-4-methylbenzene **2b** (0.6 mmol, 90.4 mg), $CoFe_2O_4$ (0.02 mmol, 5.2 mg), DABCO (0.4 mmol, 45.6 mg), and DMF (0.3 mL) were used. After column chromatography (hexanes/toluene/ethyl acetate 1:1:2), 46.0 mg (81% yield) of **3ab** was obtained as a yellow solid. R_f = 0.24 (hexanes/ethyl acetate 2:1), mp 212 – 214 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.20 (s, 1H), 7.65 (d, J = 8.4 Hz, 2H), 7.51 (d, J = 8.7 Hz, 1H), 7.15 (d, J = 8.3 Hz, 2H), 6.93 (d, J = 2.4 Hz, 1H), 6.63 (dd, J = 8.7, 2.5 Hz, 1H), 2.91 (s, 6H), 2.27 (s, 3H). ^{13}C NMR (150 MHz, DMSO-d_6) δ 162.1, 153.6, 149.9, 138.4, 130.7, 129.3, 120.8, 117.7, 117.2, 109.0, 103.3, 40.7, 20.4. HRMS (ESI) *m*/*z* calcd for $C_{16}H_{17}N_3S$ $[M + H]^+$: 284.1216, found: 284.1214.

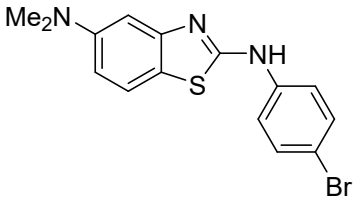
N^2-(4-Methoxyphenyl)-N^5,N^5-dimethylbenzo[*d*]thiazole-2,5-diamine (3ac)



Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.4 mg), 1-isothiocyanato-4-methoxybenzene **2c** (0.6 mmol, 112.5 mg), $CoFe_2O_4$ (0.02 mmol, 4.9 mg), DABCO (0.4 mmol,

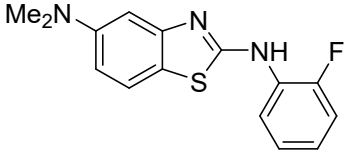
44.9 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/dichloromethane/ethyl acetate 1:1:1), 54.0 mg (90% yield) of **3ac** was obtained as an orange solid. R_f = 0.34 (hexanes/ethyl acetate 3:1), mp 215–216 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.11 (s, 1H), 7.69 – 7.64 (m, 2H), 7.49 (d, J = 8.7 Hz, 1H), 6.96 – 6.91 (m, 2H), 6.90 (d, J = 2.4 Hz, 1H), 6.61 (dd, J = 8.7, 2.5 Hz, 1H), 3.74 (s, 3H), 2.91 (s, 6H). ^{13}C NMR (150 MHz, DMSO-d_6) δ 162.5, 154.5, 153.7, 149.8, 134.3, 120.7, 119.4, 117.2, 114.2, 108.8, 103.2, 55.2, 40.7. HRMS (ESI) m/z calcd for $C_{16}H_{17}N_3OS$ $[M+H]^+$: 300.1165, found: 300.1152.

N^2-(4-Bromophenyl)-N^5,N^5-dimethylbenzo[*d*]thiazole-2,5-diamine (3ad)



Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.6 mg), 1-bromo-4-isothiocyanatobenzene **2d** (0.6 mmol, 129.7 mg), $CoFe_2O_4$ (0.02 mmol, 5.0 mg), DABCO (0.4 mmol, 45.8 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/dichloromethane/ethyl acetate 1:1:1), 65.4 mg (94% yield) of **3ad** was obtained as a yellow solid. R_f = 0.28 (hexanes/ethyl acetate 4:1), mp 244 – 246 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.47 (s, 1H), 7.77 (d, J = 8.8 Hz, 2H), 7.55 (d, J = 8.7 Hz, 1H), 7.52 – 7.50 (m, 2H), 6.95 (d, J = 2.3 Hz, 1H), 6.67 (dd, J = 8.7, 2.5 Hz, 1H), 2.92 (s, 6H). ^{13}C NMR (150 MHz, DMSO-d_6) δ 161.7, 153.3, 149.9, 140.1, 131.6, 120.9, 119.4, 117.2, 112.9, 109.4, 103.4, 40.6. HRMS (ESI) m/z calcd for $C_{15}H_{14}{}^{79}BrN_3S$ $[M+H]^+$: 348.0165, found: 348.0159.

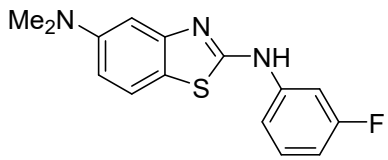
N^2-(2-Fluorophenyl)-N^5,N^5-dimethylbenzo[*d*]thiazole-2,5-diamine (3ae)



Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.4 mg), 1-fluoro-2-isothiocyanatobenzene **2e** (0.6 mmol, 92 mg), $CoFe_2O_4$ (0.02 mmol, 4.9 mg), DABCO (0.4 mmol, 46.2 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/hexanes/ethyl acetate 2:2:1), 18.5 mg (32% yield) of **3ae** was obtained as a white solid. R_f = 0.29 (hexanes/ethyl acetate 6:1), mp 188 – 190 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.13 (s, 1H), 8.53 (s, 1H), 7.54 (d, J = 8.7 Hz, 1H), 7.26 (ddd, J = 11.6, 8.1, 1.4 Hz, 1H), 7.22 (td, J = 8.0, 1.1 Hz, 1H), 7.09 – 7.03 (m, 1H), 6.93 (s, 1H), 6.67 (dd, J = 8.7, 2.4 Hz, 1H), 2.91 (s, 6H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 162.5, 152.2 (d, J = 244.8), 149.8, 124.5 (d, J = 3.1 Hz), 123.0 (d, J = 7.5

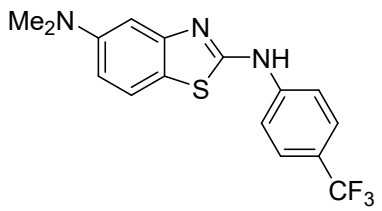
Hz), 121.3, 120.8, 115.2 (d, J = 18.9 Hz), 109.3, 103.3, 40.6. Three carbon signal could not be located. ^{19}F NMR (471 MHz, DMSO-d_6) δ -127.18 (s). HRMS (ESI) m/z calcd for $C_{15}H_{15}FN_3S$ $[M+H]^+$: 288.0965, found: 288.0972.

N^2-(3-Fluorophenyl)-N^5,N^5-dimethylbenzo[*d*]thiazole-2,5-diamine (3af)



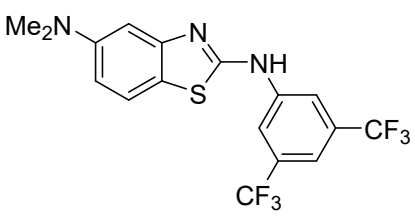
Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.5 mg), 1-fluoro-3-isothiocyanatobenzene **2f** (0.6 mmol, 92.1 mg), $CoFe_2O_4$ (0.02 mmol, 5.0 mg), DABCO (0.4 mmol, 45.3 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/hexanes/ethyl acetate 1:1:1), 24.2 mg (42% yield) of **3af** was obtained as a pale yellow solid. R_f = 0.34 (hexanes/ethyl acetate 5:1), mp 189 – 191 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.55 (s, 1H), 7.93 – 7.88 (m, 1H), 7.56 (d, J = 8.7 Hz, 1H), 7.42 – 7.37 (m, 1H), 7.36 (td, J = 8.0, 6.8 Hz, 1H), 7.00 (d, J = 2.4 Hz, 1H), 6.83 – 6.77 (m, 1H), 6.68 (dd, J = 8.7, 2.5 Hz, 1H), 2.93 (s, 6H). ^{13}C NMR (126 MHz, DMSO-d_6) δ 162.5 (d, J = 241.4 Hz), 161.6, 153.3, 149.9, 142.4 (d, J = 11.5 Hz), 130.40 (d, J = 9.8 Hz), 120.9, 117.1, 113.4, 109.5, 107.9 (d, J = 21.2 Hz), 104.3 (d, J = 27.0 Hz), 103.6, 40.6. ^{19}F NMR (471 MHz, DMSO-d_6) δ -111.80 (s). HRMS (ESI) m/z calcd for $C_{15}H_{15}FN_3S$ $[M+H]^+$: 288.0965, found: 288.0953.

N^5,N^5-Dimethyl-N^2-(4-(trifluoromethyl)phenyl)benzo[*d*]thiazole-2,5-diamine (3ag)



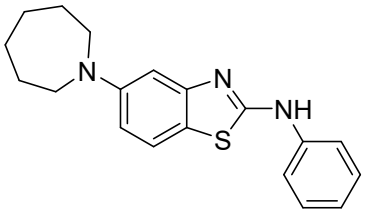
Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.4 mg), 1-isothiocyanato-4-(trifluoromethyl)benzene **2g** (0.6 mmol, 122.5 mg), $CoFe_2O_4$ (0.02 mmol, 5.0 mg), DABCO (0.4 mmol, 45.2 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/hexanes/ethyl acetate 1:1:1), 39.0 mg (58% yield) of **3ag** was obtained as a yellow solid. R_f = 0.29 (hexanes/ethyl acetate 4:1), mp 226 – 228 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.73 (s, 1H), 7.98 (d, J = 8.6 Hz, 2H), 7.70 (d, J = 8.6 Hz, 2H), 7.59 (d, J = 8.7 Hz, 1H), 6.99 (d, J = 2.4 Hz, 1H), 6.71 (dd, J = 8.8, 2.5 Hz, 1H), 2.93 (s, 6H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 161.5, 153.1, 149.9, 144.1, 126.2 (q, J = 3.4 Hz), 121.0, 117.3, 117.2, 109.7, 103.5, 40.6. ^{19}F NMR (471 MHz, DMSO-d_6) δ -59.88 (s). HRMS (ESI) m/z calcd for $C_{16}H_{15}F_3N_3S$ $[M+H]^+$: 338.0933, found: 338.0934.

N^2-(3,5-bis(Trifluoromethyl)phenyl)-N^5,N^5-dimethylbenzo[*d*]thiazole-2,5-diamine (3ah)



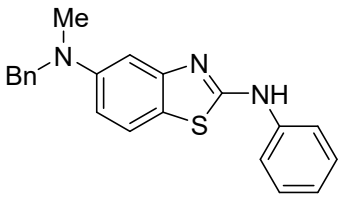
Following the general procedure, in which *N*,*N*-dimethyl-3-nitroaniline **1a** (0.2 mmol, 33.3 mg), 1-isothiocyanato-3,5-bis(trifluoromethyl)benzene **2h** (0.6 mmol, 162.8 mg), $CoFe_2O_4$ (0.02 mmol, 4.9 mg), DABCO (0.4 mmol, 44.9 mg), and DMF (0.3 mL) were used. After column chromatography (hexanes/ethyl acetate 5:1), 65.7 mg (81% yield) of **3ah** was obtained as a yellow solid. R_f = 0.31 (hexanes/ethyl acetate 8:1), mp 146 – 148 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 11.04 (s, 1H), 8.44 (s, 2H), 7.64 (s, 1H), 7.61 (d, J = 8.7 Hz, 1H), 6.98 (d, J = 2.4 Hz, 1H), 6.73 (dd, J = 8.8, 2.5 Hz, 1H), 2.94 (s, 6H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 161.3, 152.9, 150.0, 142.3, 130.9 (q, J = 33.0 Hz), 123.3 (q, J = 273.1 Hz), 121.1, 117.1, 116.9 (q, J = 3.2 Hz), 114.1 – 113.7 (m), 110.0, 103.6, 40.6. One carbon signal could not be located. ^{19}F NMR (471 MHz, DMSO-d_6) δ -61.63 (s). HRMS (ESI) *m*/*z* calcd for $C_{17}H_{14}F_6N_3S$ $[M+H]^+$: 406.0807, found: 406.0797.

5-(Azepan-1-yl)-*N*-phenylbenzo[*d*]thiazol-2-amine (3ba)



Following the general procedure, in which 1-(3-nitrophenyl)azepane **1b** (0.2 mmol, 44.6 mg), phenyl isothiocyanate **2a** (0.6 mmol, 81.2 mg), $CoFe_2O_4$ (0.02 mmol, 4.8 mg), DABCO (0.4 mmol, 44.9 mg) and DMF (0.3 mL) were used. After column chromatography (hexanes/ethyl acetate 4:1) 36.3 mg (56% yield) of **3ba** was obtained as an orange solid. R_f = 0.29 (hexanes/ethyl acetate 6:1), mp 159 – 161 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.27 (s, 1H), 7.77 (d, J = 7.9 Hz, 2H), 7.47 (d, J = 8.7 Hz, 1H), 7.34 (dd, J = 8.4, 7.5 Hz, 2H), 6.99 (t, J = 7.3 Hz, 1H), 6.88 (d, J = 2.3 Hz, 1H), 6.59 (dd, J = 8.8, 2.4 Hz, 1H), 3.48 (t, J = 6.0 Hz, 4H), 1.75 (s, 4H), 1.49 – 1.44 (m, 4H). ^{13}C NMR (150 MHz, DMSO-d_6) δ 161.9, 153.9, 147.6, 140.9, 128.9, 121.7, 121.1, 117.6, 115.6, 107.7, 101.8, 49.1, 27.0, 26.5. HRMS (ESI) *m*/*z* calcd for $C_{19}H_{22}N_3S$ $[M+H]^+$: 324.1529, found: 324.1517.

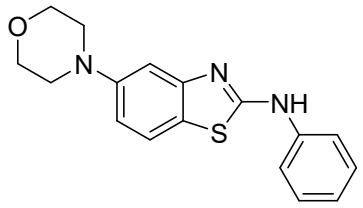
N^5-Benzyl-N^5-methyl-N^2-phenylbenzo[*d*]thiazole-2,5-diamine (3ca)



Following the general procedure, in which *N*-benzyl-*N*-methyl-3-nitroaniline **1c** (0.2 mmol, 49.1 mg), phenyl

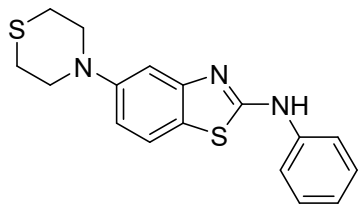
isothiocyanate **2a** (0.6 mmol, 82.1 mg), $CoFe_2O_4$ (0.02 mmol, 4.7 mg), DABCO (0.4 mmol, 44.9 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/dichloromethane 1:1), 51.9 mg (75% yield) of **3ca** was obtained as an orange solid. R_f = 0.4 (hexanes/ethyl acetate 5 : 1), mp 176 – 177 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.30 (s, 1H), 7.75 (d, J = 7.9 Hz, 2H), 7.50 (d, J = 8.7 Hz, 1H), 7.34 – 7.29 (m, 4H), 7.23 – 7.20 (m, 3H), 6.98 (t, J = 7.3 Hz, 1H), 6.92 (d, J = 2.3 Hz, 1H), 6.65 (dd, J = 8.7, 2.4 Hz, 1H), 4.61 (s, 2H), 3.05 (s, 3H). ^{13}C NMR (150 MHz, DMSO-d_6) δ 162.0, 153.6, 148.5, 140.8, 139.1, 128.9, 128.4, 126.7, 126.6, 121.8, 120.9, 117.6, 117.0, 108.8, 103.0, 55.8, 39.1. HRMS (ESI) m/z calcd for $C_{21}H_{20}N_3S$ $[M+H]^+$: 346.1372, found: 346.1374.

5-Morpholino-*N*-phenylbenzo[*d*]thiazol-2-amine (3da)



Following the general procedure, in which 4-(3-nitrophenyl)morpholine **1d** (0.2 mmol, 41.8 mg), phenyl isothiocyanate **2a** (0.6 mmol, 81.8 mg), $CoFe_2O_4$ (0.02 mmol, 4.9 mg), DABCO (0.4 mmol, 45.4 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/dichloromethane/ethyl acetate 2:2:1), 52.3 mg (84% yield) of **3da** was obtained as a pale yellow solid. R_f = 0.36 (hexanes/ethyl acetate 3:2), mp 205 – 207 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.37 (s, 1H), 7.78 (d, J = 7.7 Hz, 2H), 7.60 (d, J = 8.7 Hz, 1H), 7.37 – 7.32 (m, 2H), 7.16 (d, J = 2.3 Hz, 1H), 7.03 – 6.98 (m, 1H), 6.85 (dd, J = 8.7, 2.4 Hz, 1H), 3.77 – 3.73 (m, 4H), 3.15 – 3.10 (m, 4H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 162.1, 153.3, 150.4, 140.7, 128.9, 121.8, 120.9, 120.3, 117.7, 111.7, 106.0, 66.1, 49.2. HRMS (ESI) m/z calcd for $C_{17}H_{17}N_3OS$ $[M+H]^+$: 312.1165, found: 312.1173.

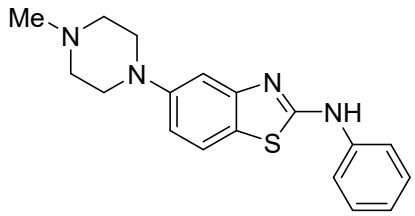
***N*-Phenyl-5-thiomorpholinobenzo[*d*]thiazol-2-amine (3ea)**



Following the general procedure, in which 4-(3-nitrophenyl)thiomorpholine **1e** (0.2 mmol, 46.0 mg), phenyl isothiocyanate **2a** (0.6 mmol, 82.0 mg), $CoFe_2O_4$ (0.02 mmol, 5.2 mg), DABCO (0.4 mmol, 45.3 mg), and DMF (0.3 mL) were used. After column chromatography (hexanes/dichloromethane 1:1), 51.6 mg (79% yield) of **3ea** was obtained as a pale yellow solid. R_f = 0.35 (hexanes/ethyl acetate 6:1), mp 215 – 217 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.36 (s, 1H), 7.78 (d,

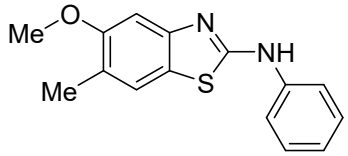
J = 7.8 Hz, 2H), 7.58 (d, *J* = 8.7 Hz, 1H), 7.37 – 7.32 (m, 2H), 7.15 (d, *J* = 2.3 Hz, 1H), 7.03 – 6.98 (m, 1H), 6.82 (dd, *J* = 8.7, 2.4 Hz, 1H), 3.55 – 3.51 (m, 4H), 2.71 – 2.67 (m, 4H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 162.1, 153.5, 150.0, 140.7, 128.9, 121.8, 121.0, 120.0, 117.7, 112.9, 107.2, 51.8, 25.7. HRMS (ESI) *m*/*z* calcd for $C_{17}H_{17}N_3S_2$ $[M+H]^+$: 328.0937, found: 328.0943.

5-(4-Methylpiperazin-1-yl)-*N*-phenylbenzo[*d*]thiazol-2-amine (3fa)



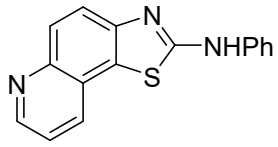
Following the general procedure, in which 1-methyl-4-(3-nitrophenyl)piperazine **1f** (0.2 mmol, 44.3 mg), phenyl isothiocyanate **2a** (0.6 mmol, 82.1 mg), $CoFe_2O_4$ (0.02 mmol, 4.9 mg), DABCO (0.4 mmol, 45.1 mg), and DMF (0.3 mL) were used. After column chromatography (hexane/ethyl acetate 1:1), 38.4 mg (59% yield) of **3fa** was obtained as a purple solid. R_f = 0.25 (toluene/ethyl acetate/methanol 10:10:3), mp 215 – 217 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.35 (s, 1H), 7.78 (dd, *J* = 8.6, 1.0 Hz, 2H), 7.57 (d, *J* = 8.7 Hz, 1H), 7.37 – 7.32 (m, 2H), 7.14 (d, *J* = 2.4 Hz, 1H), 7.02 – 6.98 (m, 1H), 6.84 (dd, *J* = 8.7, 2.4 Hz, 1H), 3.17 – 3.13 (m, 4H), 2.49 – 2.45 (m, 4H), 2.23 (s, 3H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 162.1, 153.3, 150.3, 140.7, 128.9, 121.8, 120.8, 119.9, 117.6, 112.1, 106.2, 54.6, 48.8, 45.7. HRMS (ESI) *m*/*z* calcd for $C_{18}H_{20}N_4S_2$ $[M+H]^+$: 325.1481, found: 325.1493.

5-Methoxy-6-methyl-*N*-phenylbenzo[*d*]thiazol-2-amine (3ga)



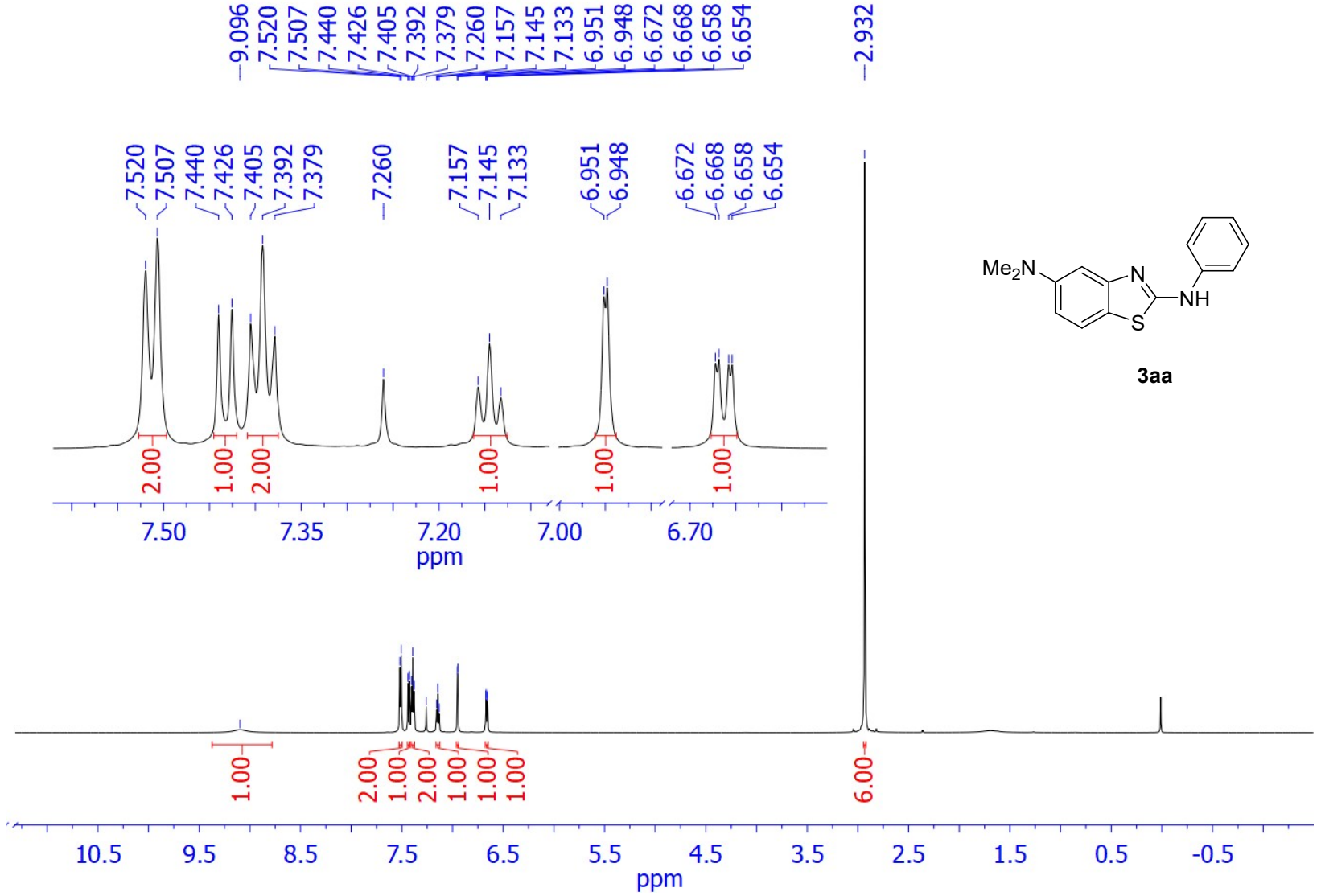
Following the general procedure, in which 2-methoxy-1-methyl-4-nitrobenzene **1g** (0.2 mmol, 33.5 mg), phenyl isothiocyanate **2a** (0.6 mmol, 82.0 mg), $CoFe_2O_4$ (0.02 mmol, 4.8 mg), DABCO (0.4 mmol, 46.9 mg), and DMF (0.3 mL) were used. After column chromatography (toluene/dichloromethane/ethyl acetate 1:1:0.1), 11.1 mg (20% yield) of **3ga** was obtained as a pale yellow solid. R_f = 0.28 (hexanes/ethyl acetate 5:1), mp 199 – 201 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.34 (s, 1H), 7.78 (d, *J* = 7.7 Hz, 2H), 7.51 (d, *J* = 0.6 Hz, 1H), 7.37 – 7.32 (m, 2H), 7.20 (s, 1H), 7.02 – 6.97 (m, 1H), 3.84 (s, 3H), 2.19 (s, 3H). ^{13}C NMR (150 MHz, DMSO-d_6) δ 161.8, 156.5, 151.5, 140.8, 128.9, 121.8, 121.5, 121.0, 120.5, 117.6, 101.7, 55.5, 16.2. HRMS (ESI) *m*/*z* calcd for $C_{15}H_{14}N_2OS$ $[M+H]^+$: 271.0900, found: 271.0910.

***N*-phenylthiazolo[4,5-*g*]quinolin-2-amine (3ha)**



Following the general procedure, in which 6-nitroquinoline **1h** (0.2 mmol, 35.1 mg), phenyl isothiocyanate **2a** (0.6 mmol, 82.0 mg), $CoFe_2O_4$ (0.02 mmol, 4.8 mg), DABCO (0.4 mmol, 45.5 mg), and DMF (0.3 mL) were used. After column chromatography (hexane/ethyl acetate/methanol 1:1:0.05), 22.8 mg (41% yield) of **3ha** was obtained as a pale yellow solid. R_f = 0.23 (hexanes/ethyl acetate 2:1), mp 245 – 247 °C. ^{1}H NMR (600 MHz, DMSO-d_6) δ 10.71 (s, 1H), 8.85 (dd, J = 4.2, 1.5 Hz, 1H), 8.34 (d, J = 8.1 Hz, 1H), 8.03 (d, J = 8.9 Hz, 1H), 7.96 (d, J = 8.9 Hz, 1H), 7.82 (d, J = 7.8 Hz, 2H), 7.56 (dd, J = 8.3, 4.2 Hz, 1H), 7.40 (dd, J = 8.2, 7.5 Hz, 2H), 7.05 (t, J = 7.3 Hz, 1H). ^{13}C NMR (125 MHz, DMSO-d_6) δ 162.7, 149.9, 148.4, 144.6, 140.5, 132.2, 129.0, 127.6, 124.1, 122.8, 122.6, 122.2, 121.7, 117.8. HRMS (ESI) m/z calcd for $C_{16}H_{12}N_3S$ $[M+H]^+$: 278.0746, found: 278.0739.

4.3. Copies of NMR spectra of products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6.654
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1.00
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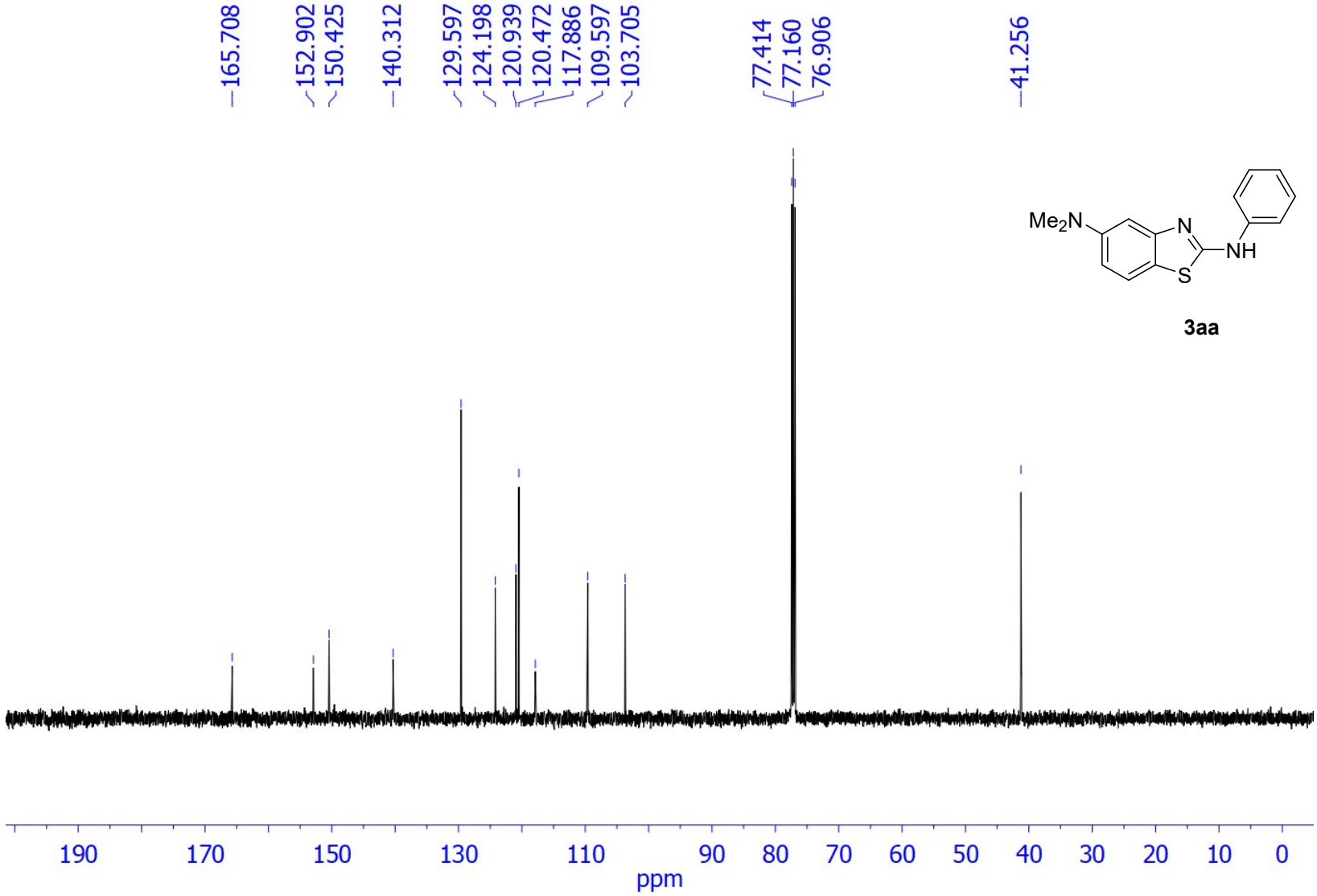
N
NH
S

3aa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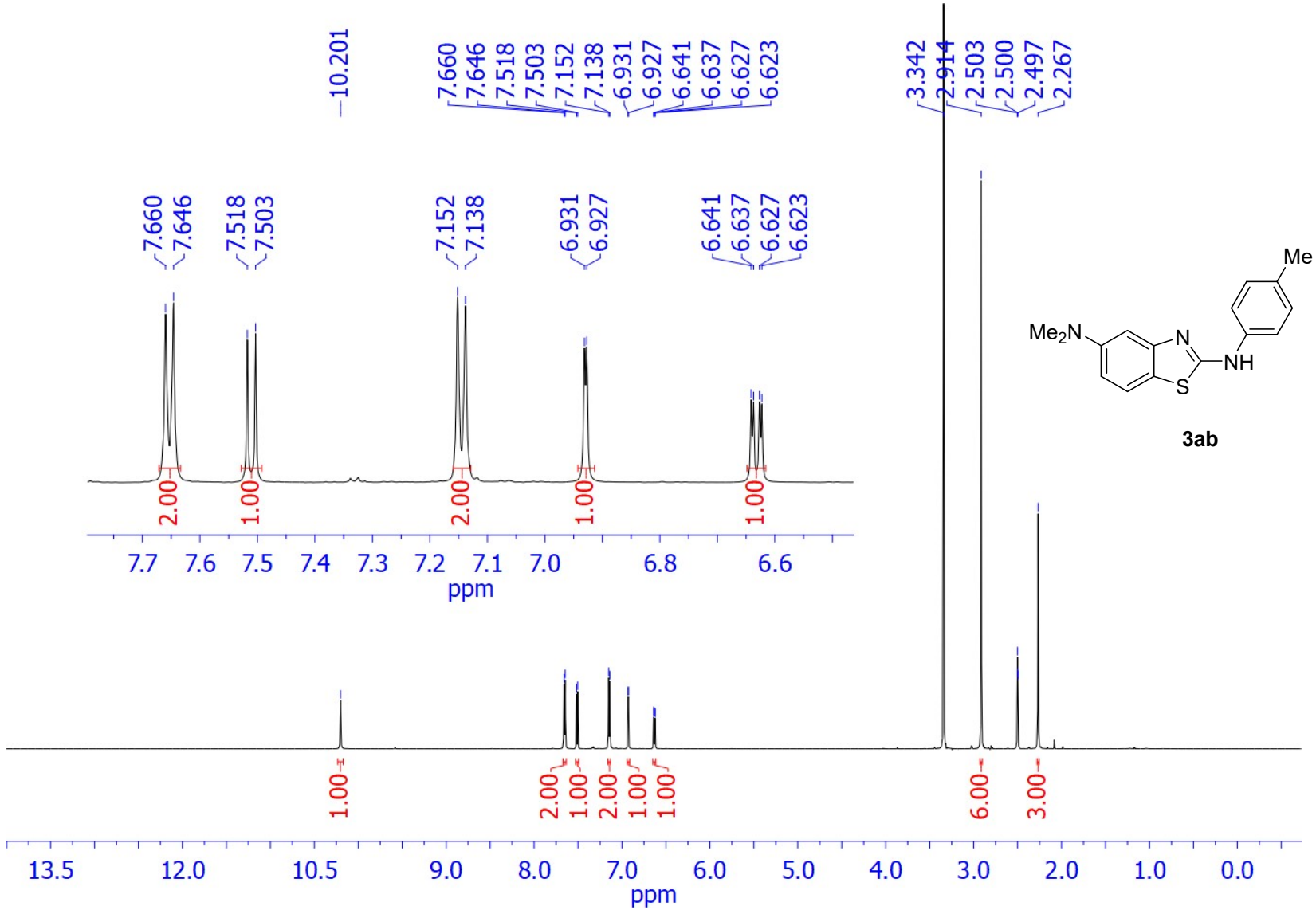
ppm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ppm
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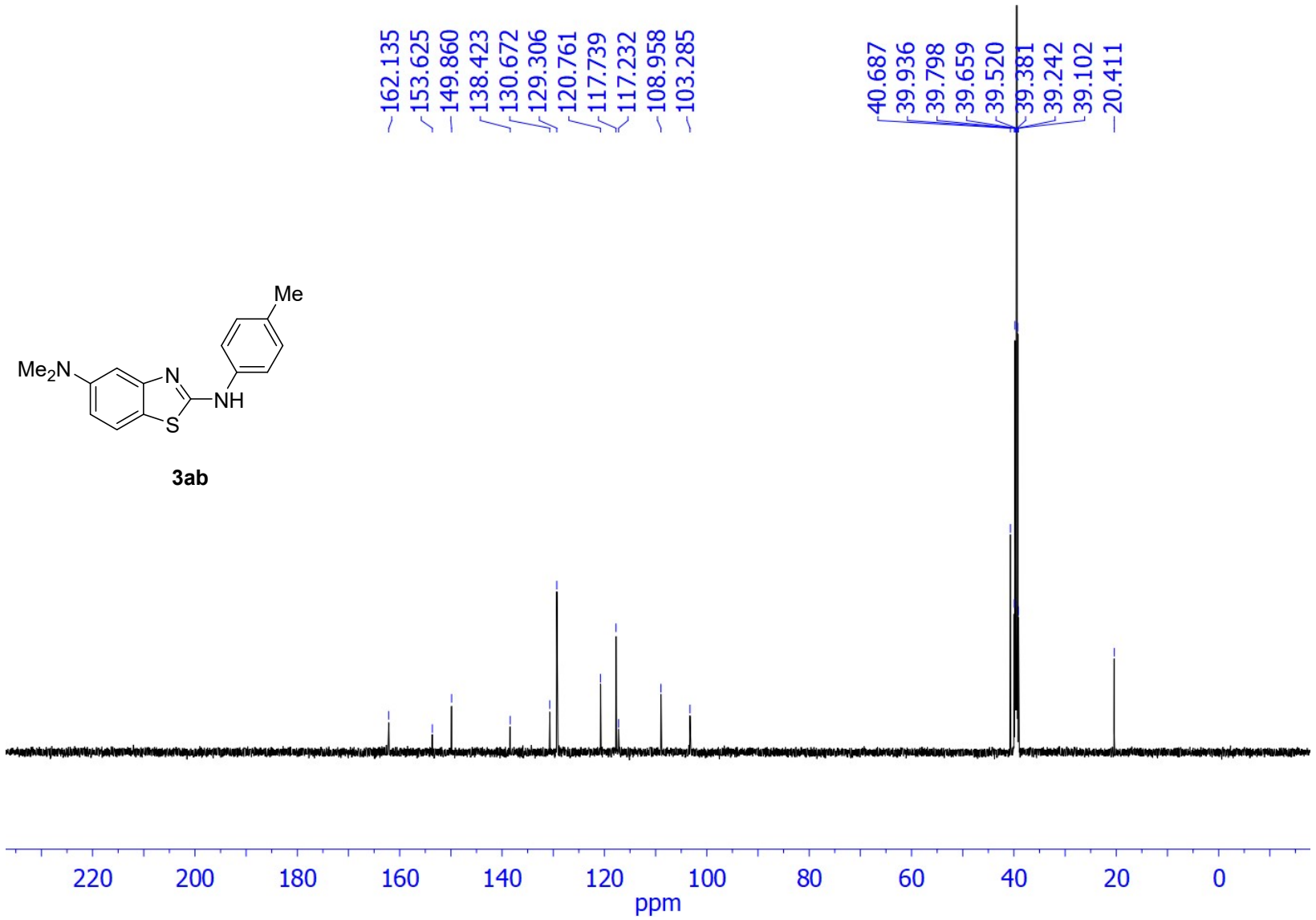
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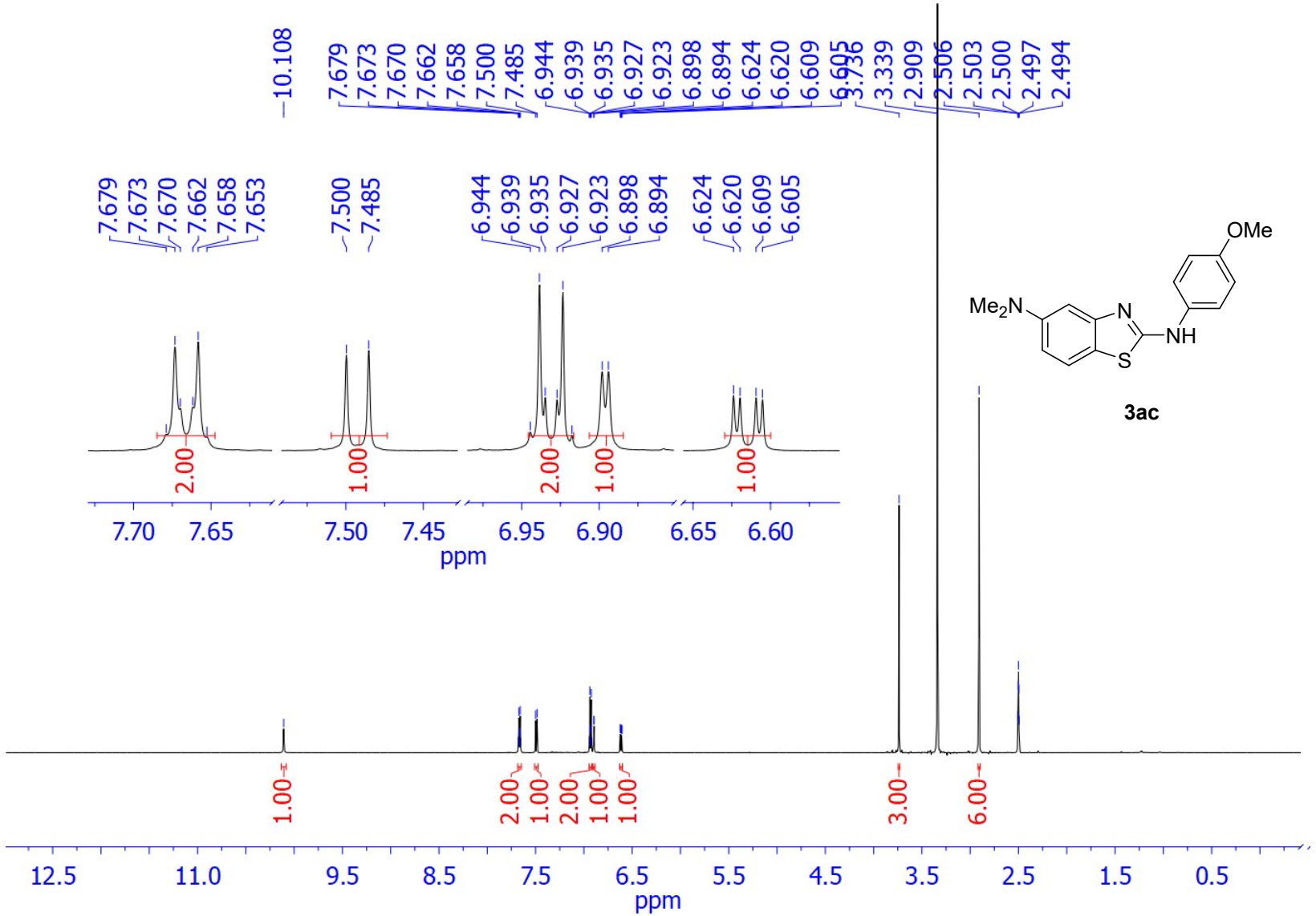
3ab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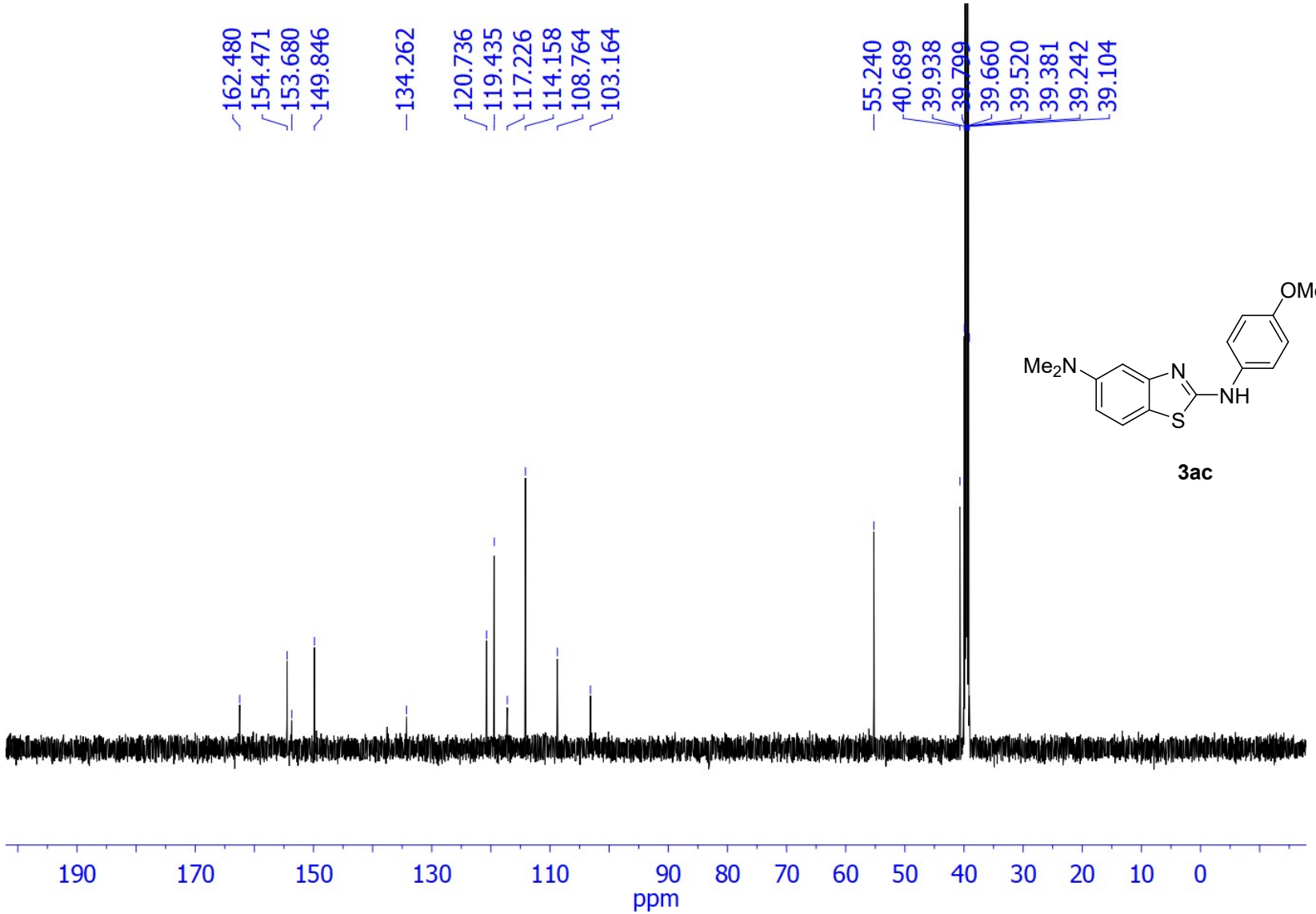
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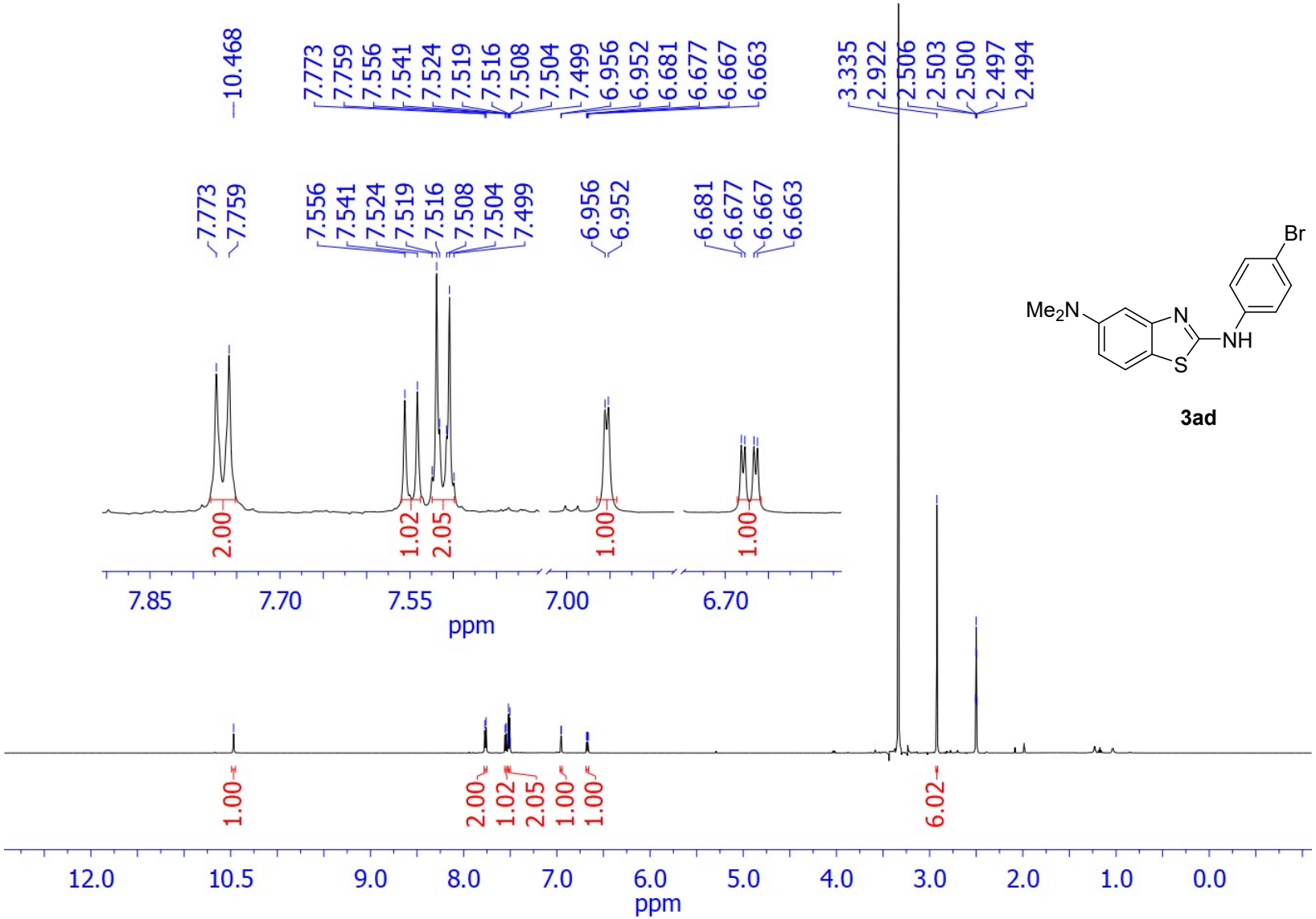
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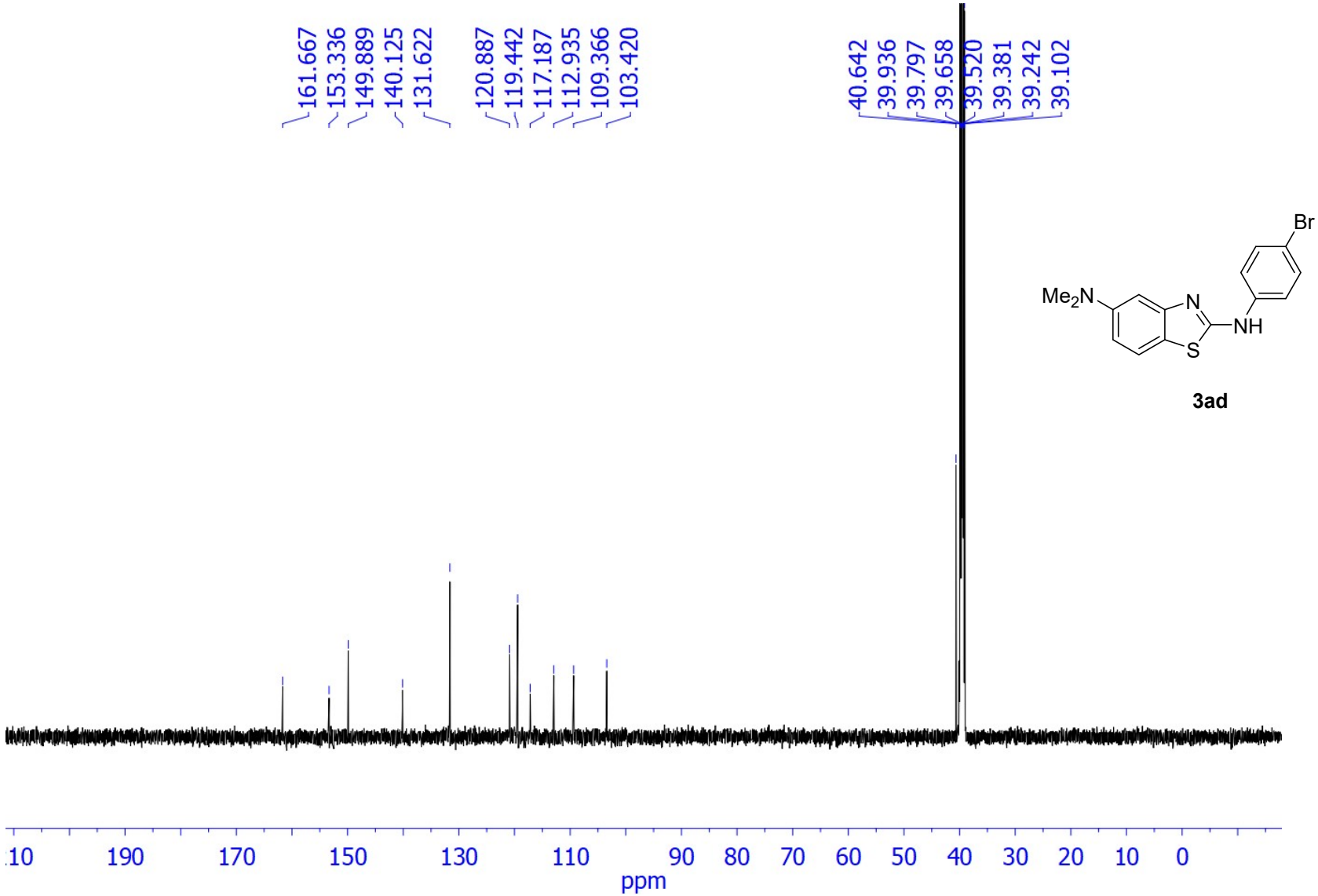
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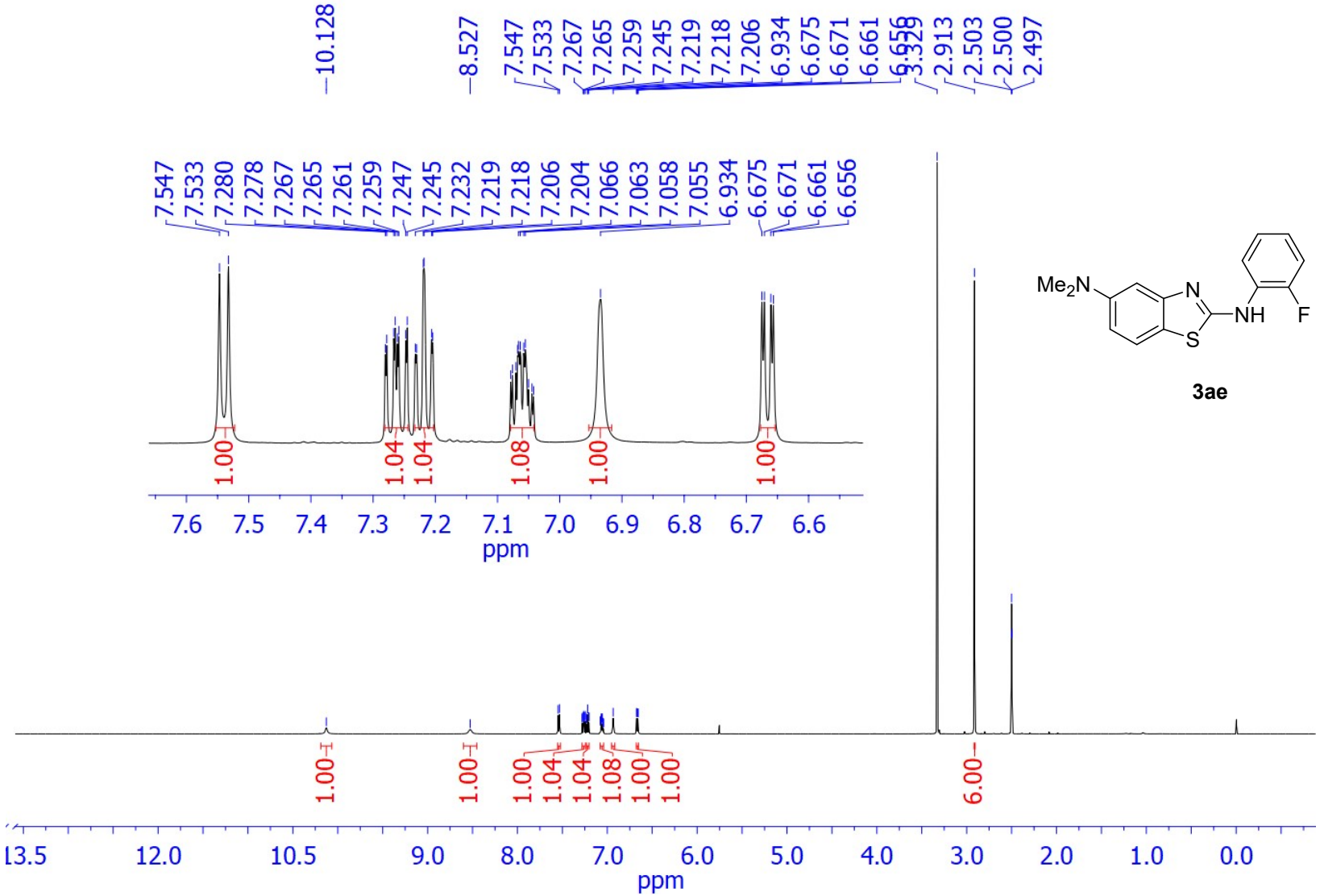
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7.0
6.9
6.8
6.7
6.6
ppm
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1.00
1.00
1.04
1.04
1.08
1.00
1.00
6.00
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12.0
10.5
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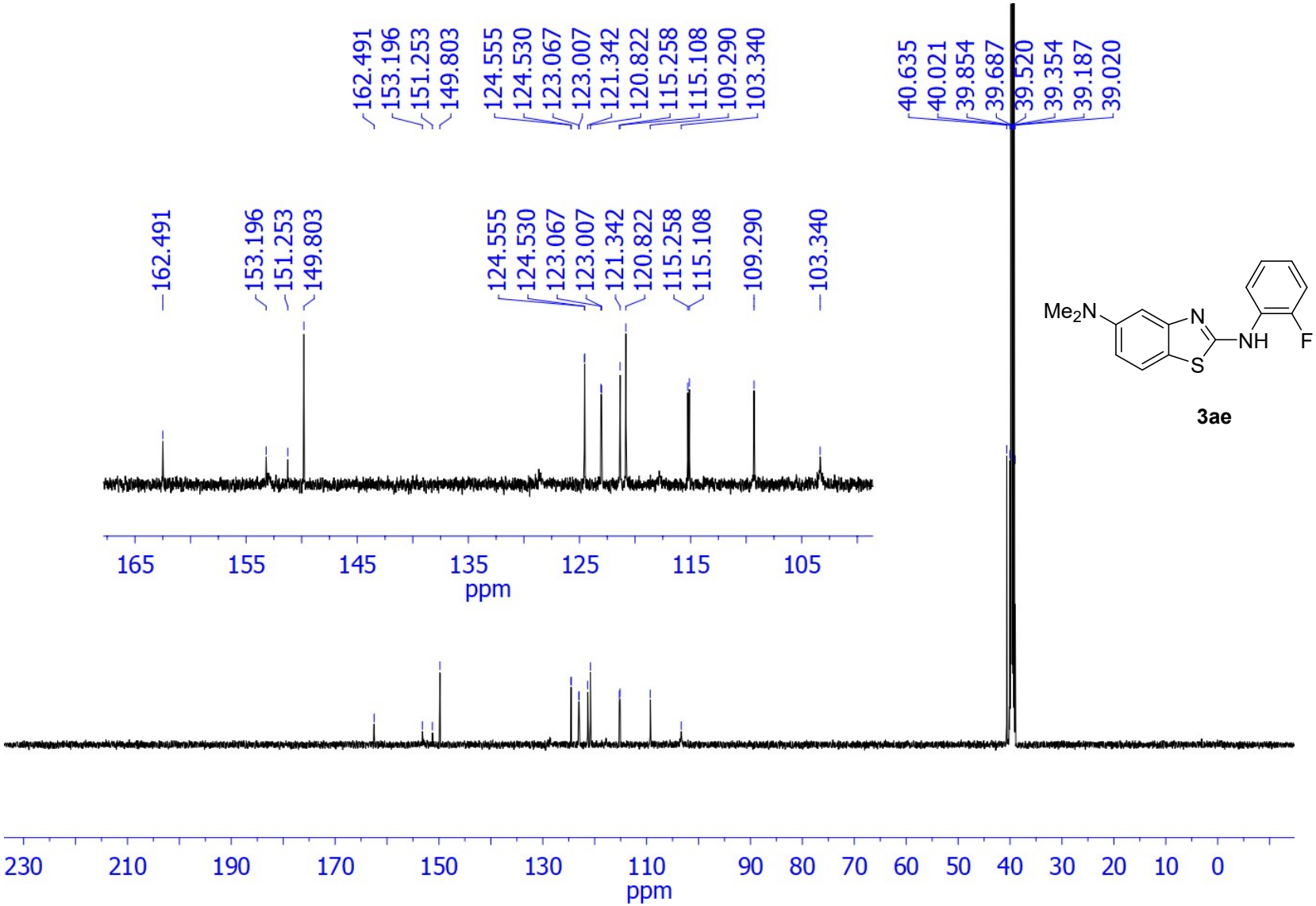
ppm
Me2N
N
NH
F
S
3ae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103.340
40.635
40.021
39.854
39.687
39.520
39.354
39.187
39.020
Me2N
N
NH
F
S
3ae
165
155
145
135
125
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ppm
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190
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150
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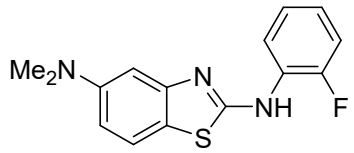
Me2N
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F
S

3ae

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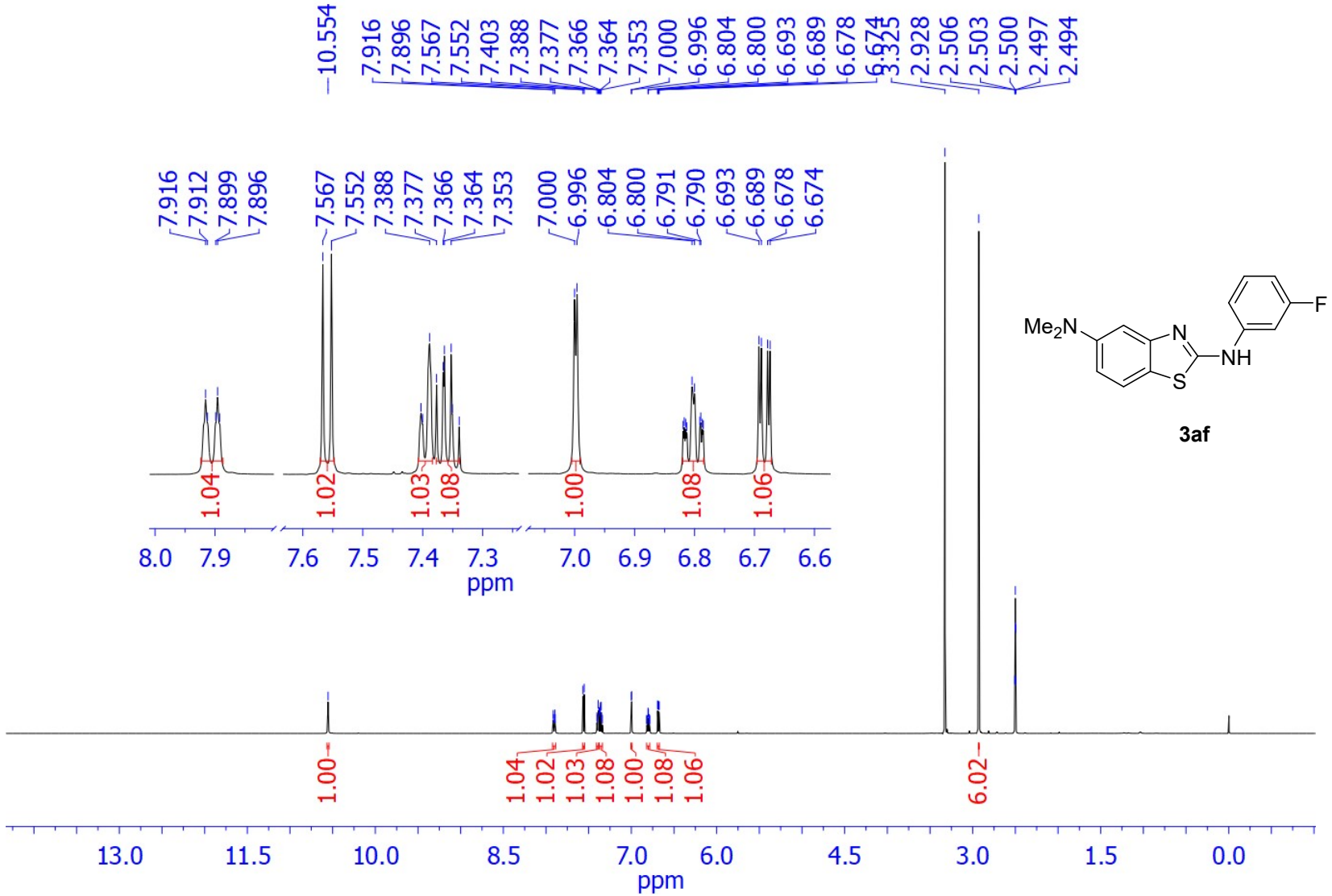
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6.996
6.804
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6.693
6.689
6.678
6.674
3.325
2.928
2.506
2.503
2.500
2.497
2.494
1.04
1.02
1.03
1.08
1.00
1.08
1.06
8.0
7.9
7.6
7.5
7.4
7.3
7.0
6.9
6.8
6.7
6.6
ppm
1.00
1.04
1.02
1.03
1.08
1.00
1.08
1.06
6.02
13.0
11.5
10.0
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4.5
3.0
1.5
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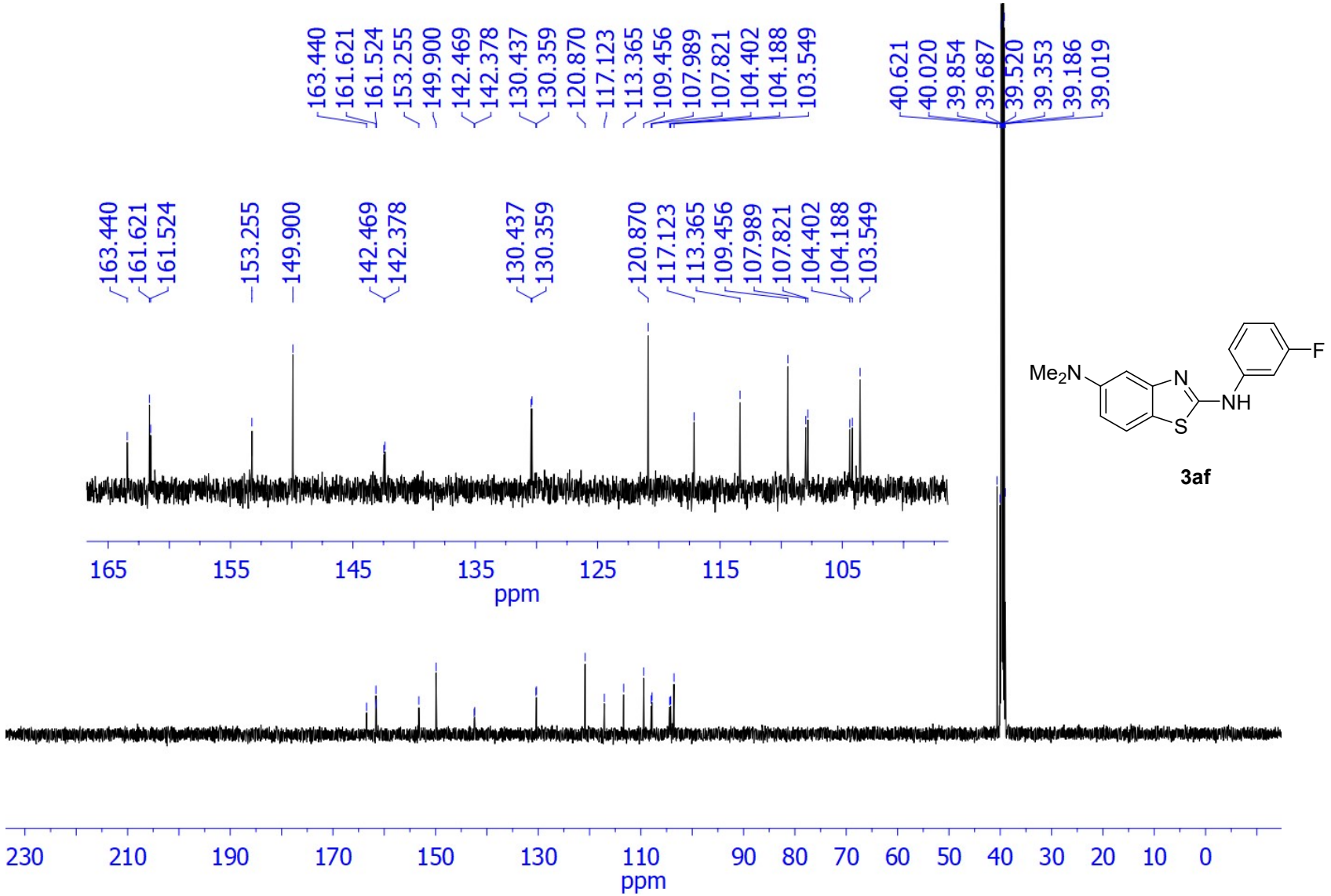
ppm
Me2N
N
NH
S
F
3af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107.821
104.402
104.188
103.549
40.621
40.020
39.854
39.687
39.520
39.353
39.186
39.019
Me2N
N
NH
S
F
3af
165
155
145
135
125
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ppm
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190
170
150
130
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60
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0
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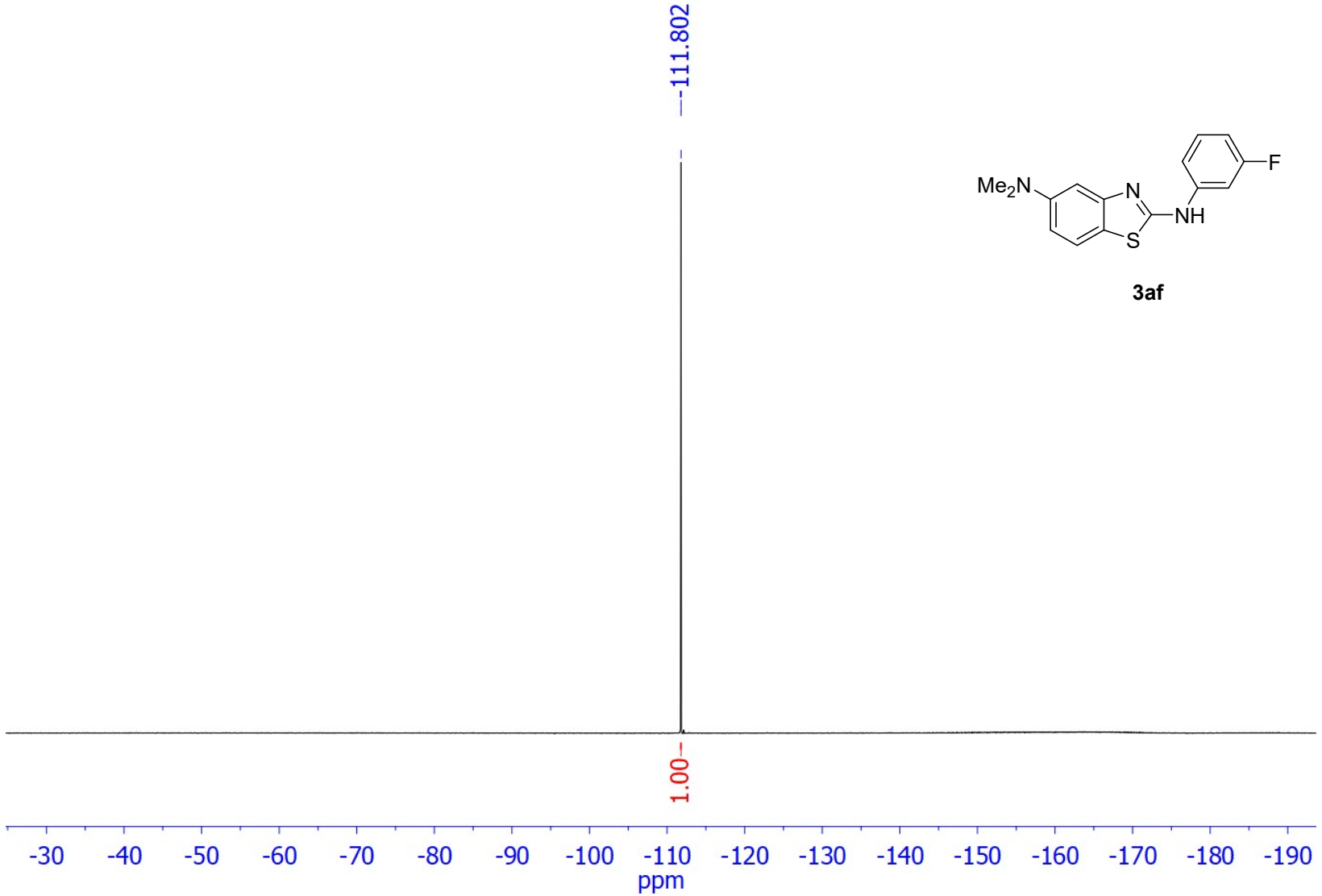
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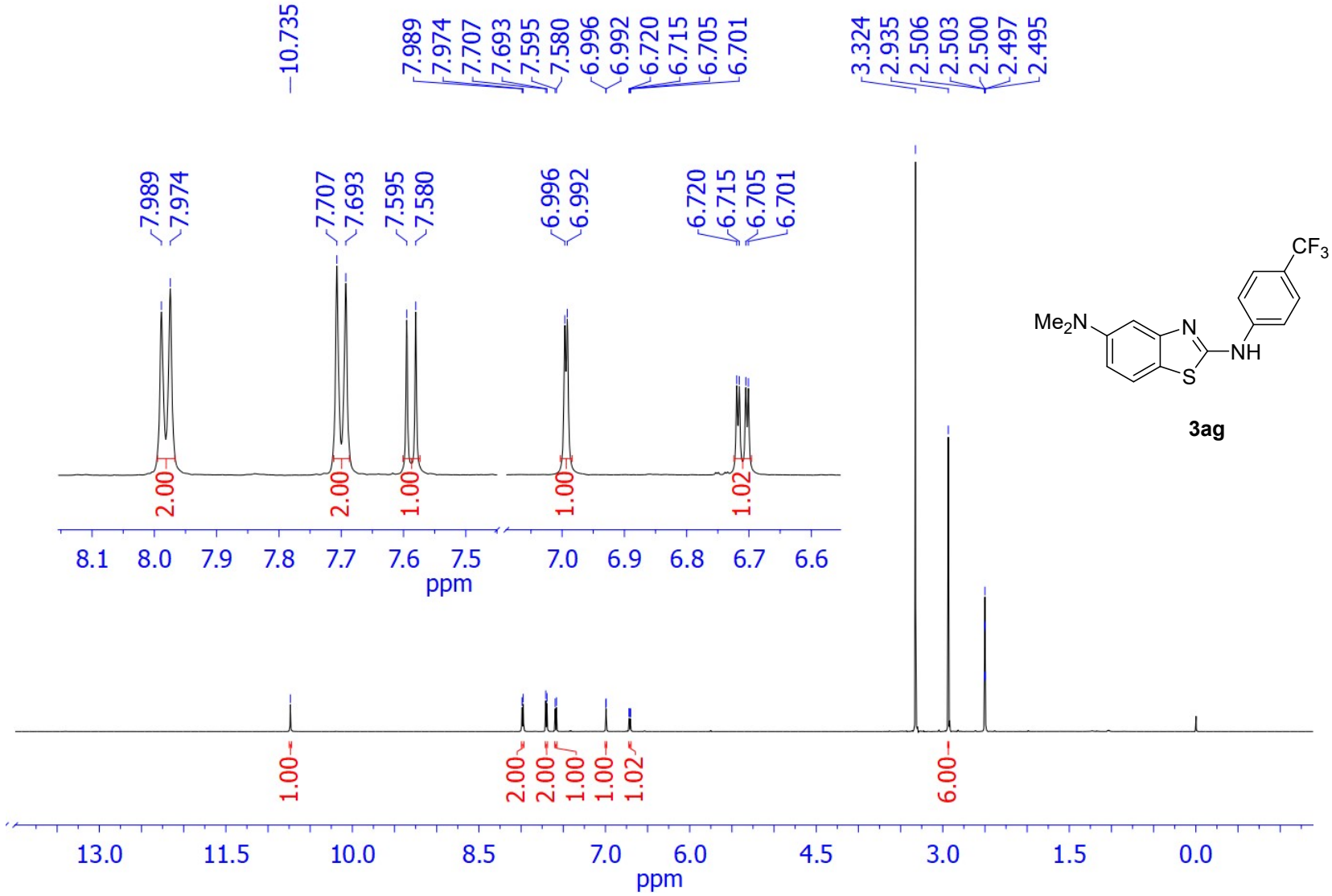
ppm

Me2N
N
NH
S
F

3af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6.996
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6.720
6.715
6.705
6.701
3.324
2.935
2.506
2.503
2.500
2.497
2.495
2.00
2.00
1.00
1.00
1.02
8.1
8.0
7.9
7.8
7.7
7.6
7.5
7.0
6.9
6.8
6.7
6.6
ppm
1.00
2.00
2.00
1.00
1.00
1.02
6.00
13.0
11.5
10.0
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4.5
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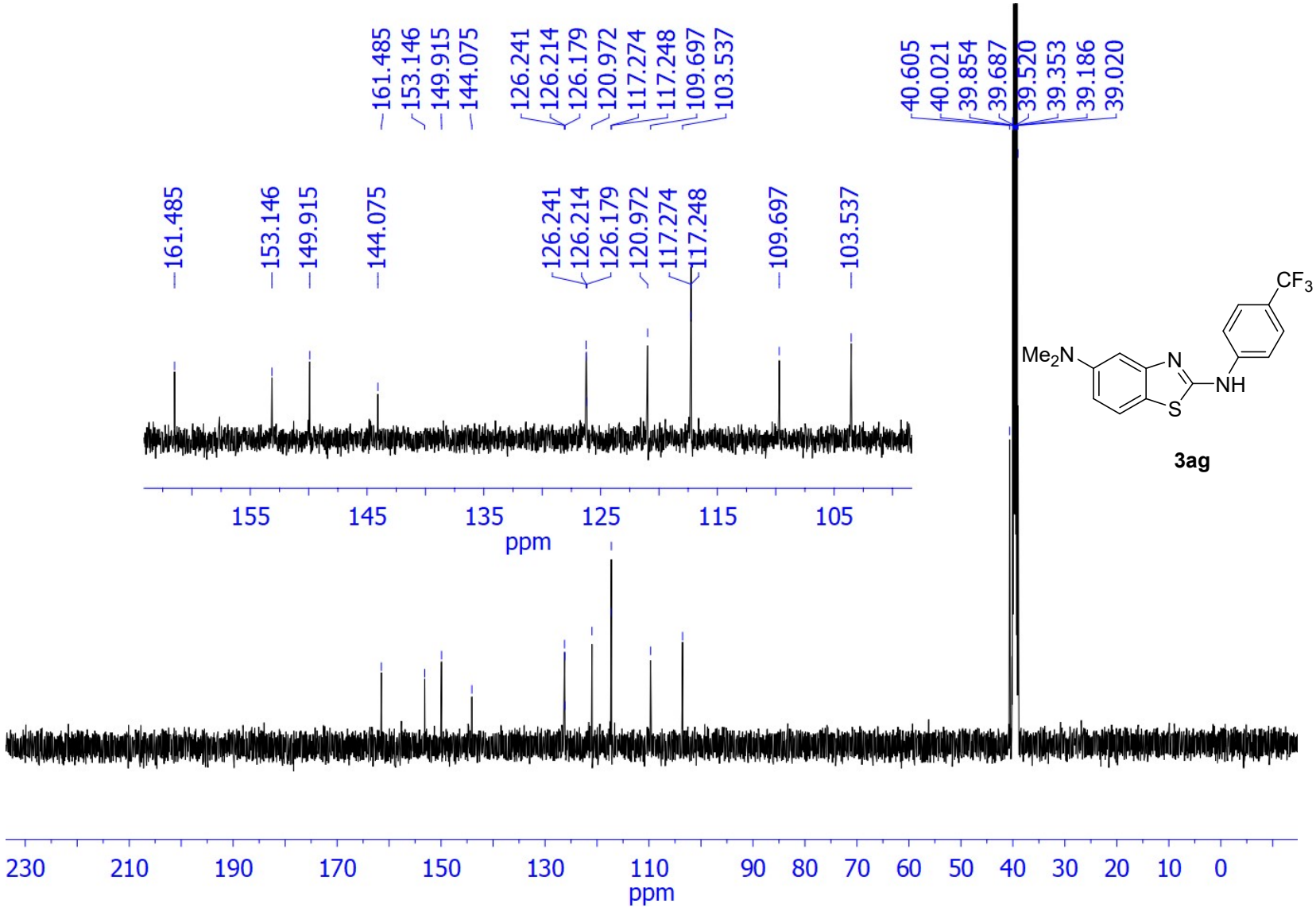
ppm
Me2N
N
NH
S
CF3

3ag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126.214
126.179
120.972
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117.248
109.697
103.537
40.605
40.021
39.854
39.687
39.520
39.353
39.186
39.020
155
145
135
125
115
105
ppm
230
210
190
170
150
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90
80
70
60
50
40
30
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10
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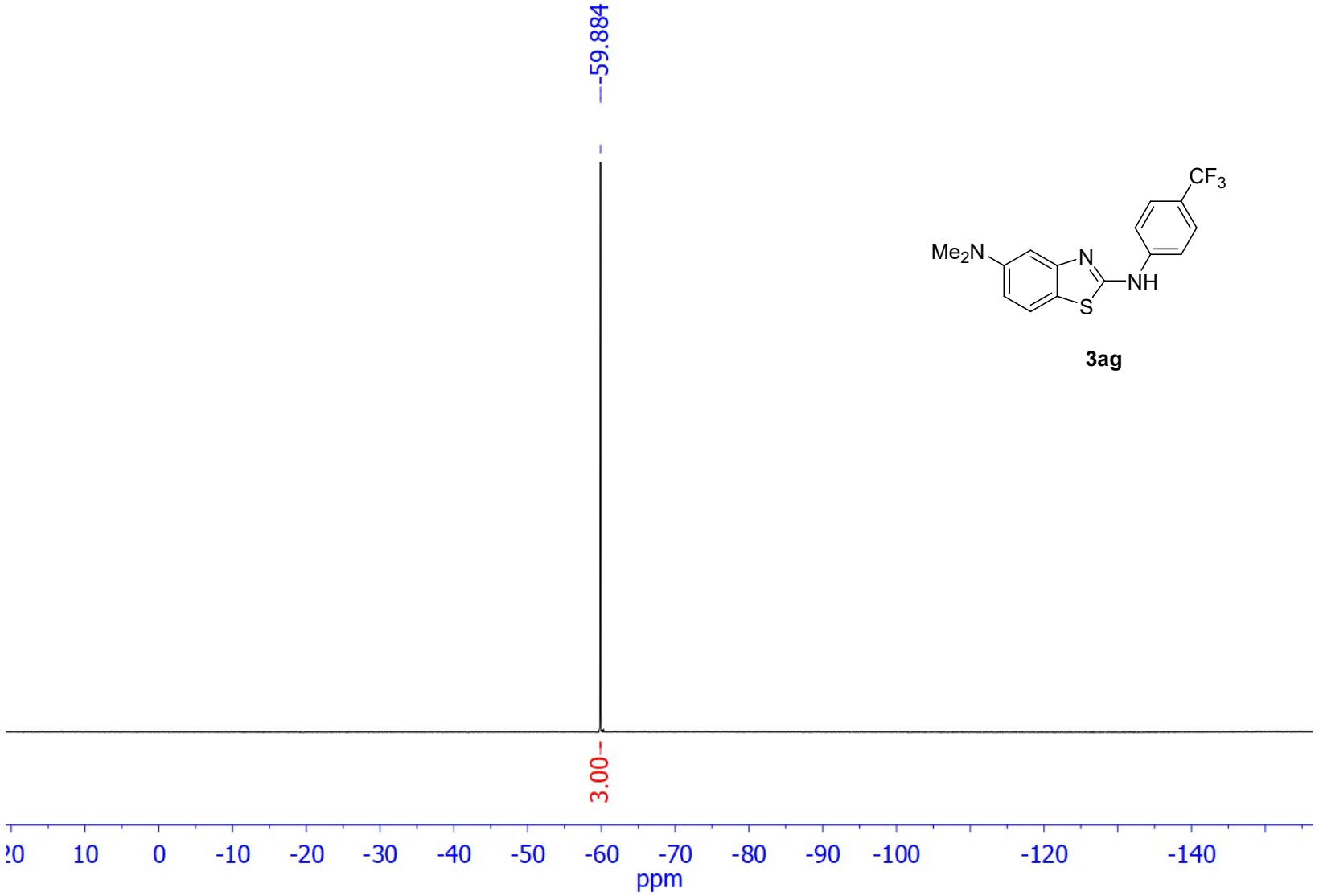
ppm
Me2N
N
NH
S
CF3
3ag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-90
-100
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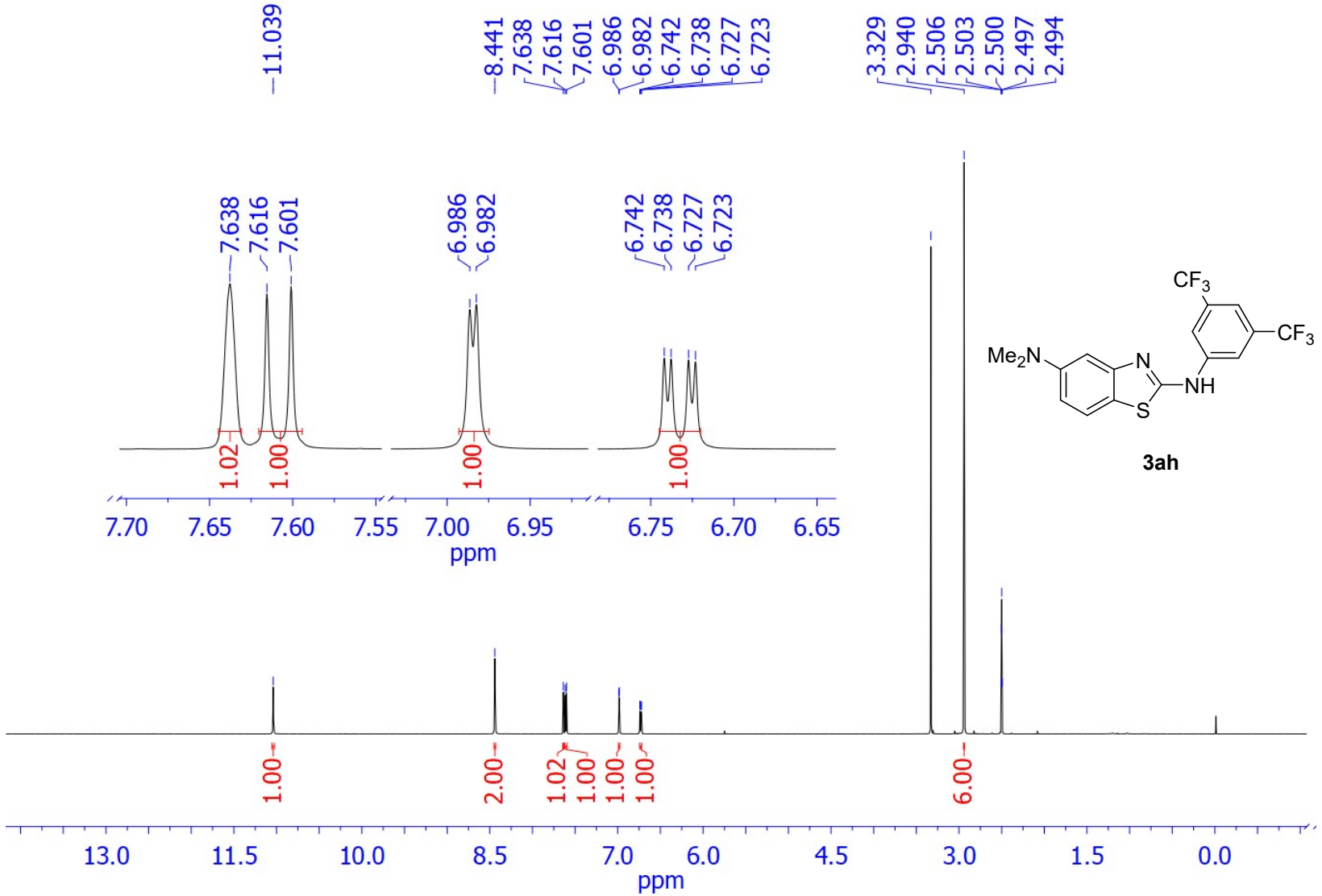
-140
ppm

Me2N
N
NH
S
CF3

3ag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6.727
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3.329
2.940
2.506
2.503
2.500
2.497
2.494
1.00
2.00
1.02
1.00
1.00
1.00
6.00
13.0
11.5
10.0
8.5
7.0
6.0
4.5
3.0
1.5
0.0
ppm
1.02
1.00
1.00
1.00
7.70
7.65
7.60
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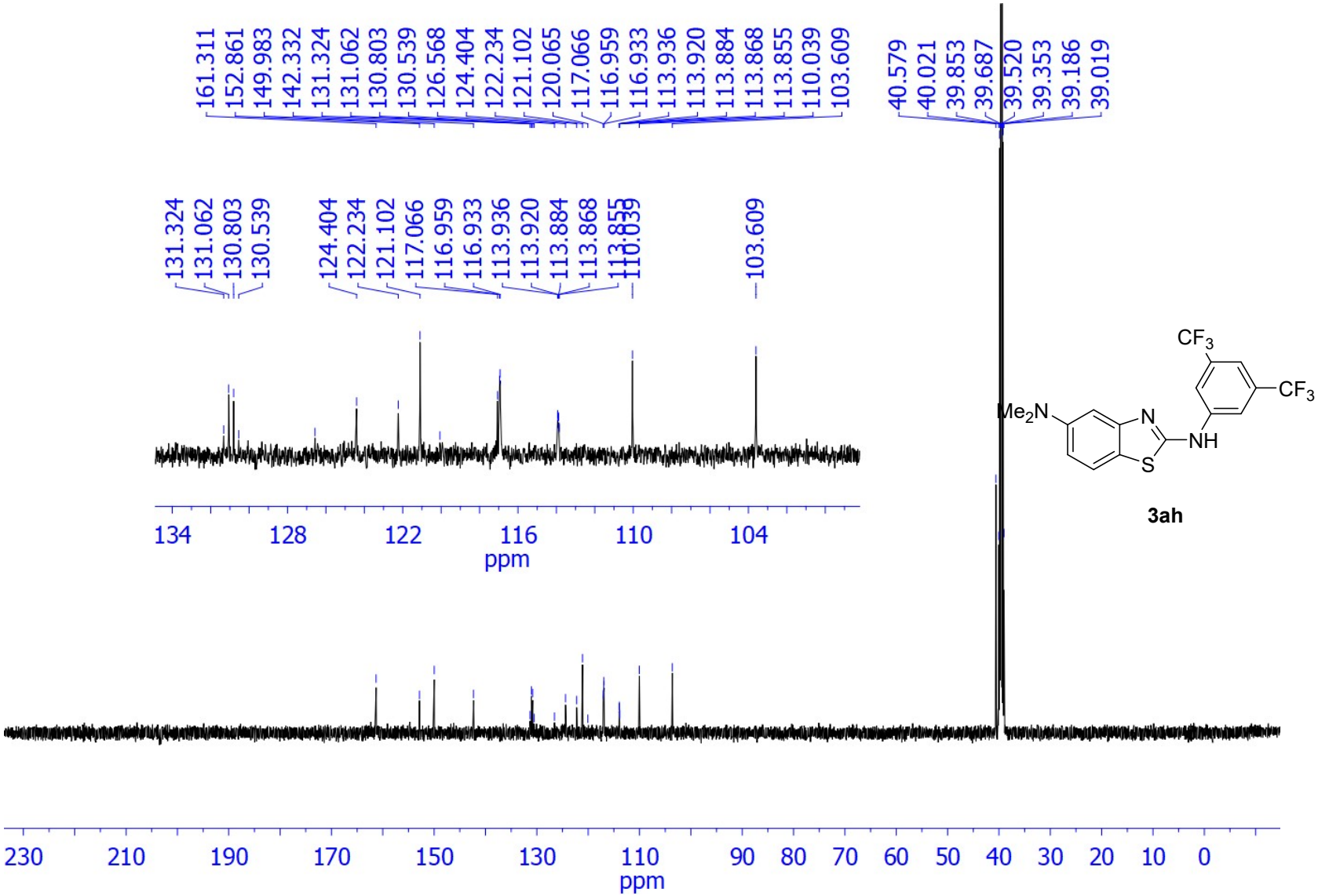
6.95
6.75
6.70
6.65
ppm
CF3
CF3
Me2N
N
NH
S

3ah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130.539
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122.234
121.102
120.065
117.066
116.959
116.933
113.936
113.920
113.884
113.868
113.855
110.039
103.609
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40.021
39.853
39.687
39.520
39.353
39.186
39.019
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131.062
130.803
130.539
124.404
122.234
121.102
117.066
116.959
116.933
113.936
113.920
113.884
113.868
113.855
110.039
103.609
134
128
122
116
110
104
ppm
CF3
CF3
Me2N
N
NH
S
3ah
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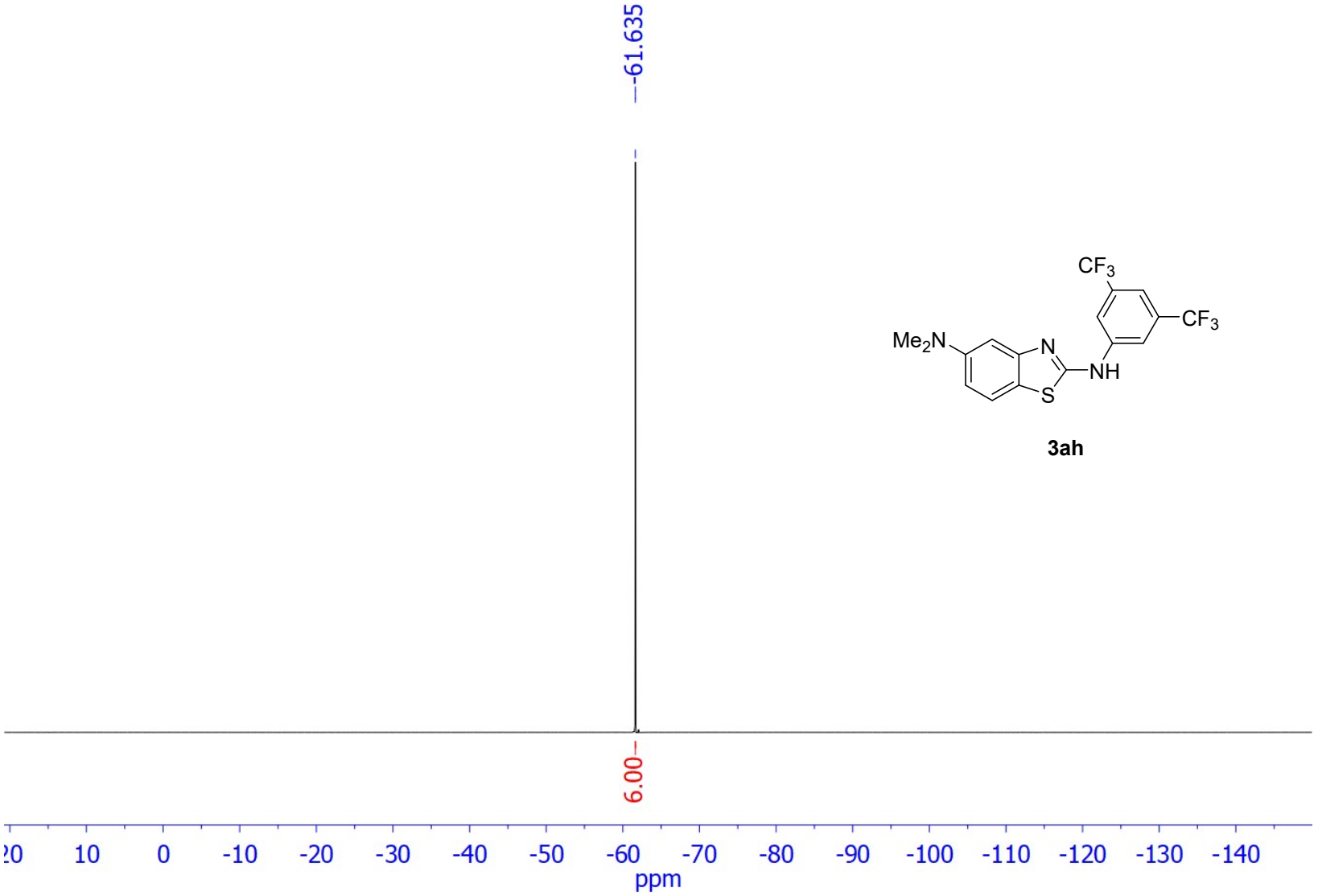
210
190
170
150
130
110
90
80
70
60
50
40
30
20
10
0
ppm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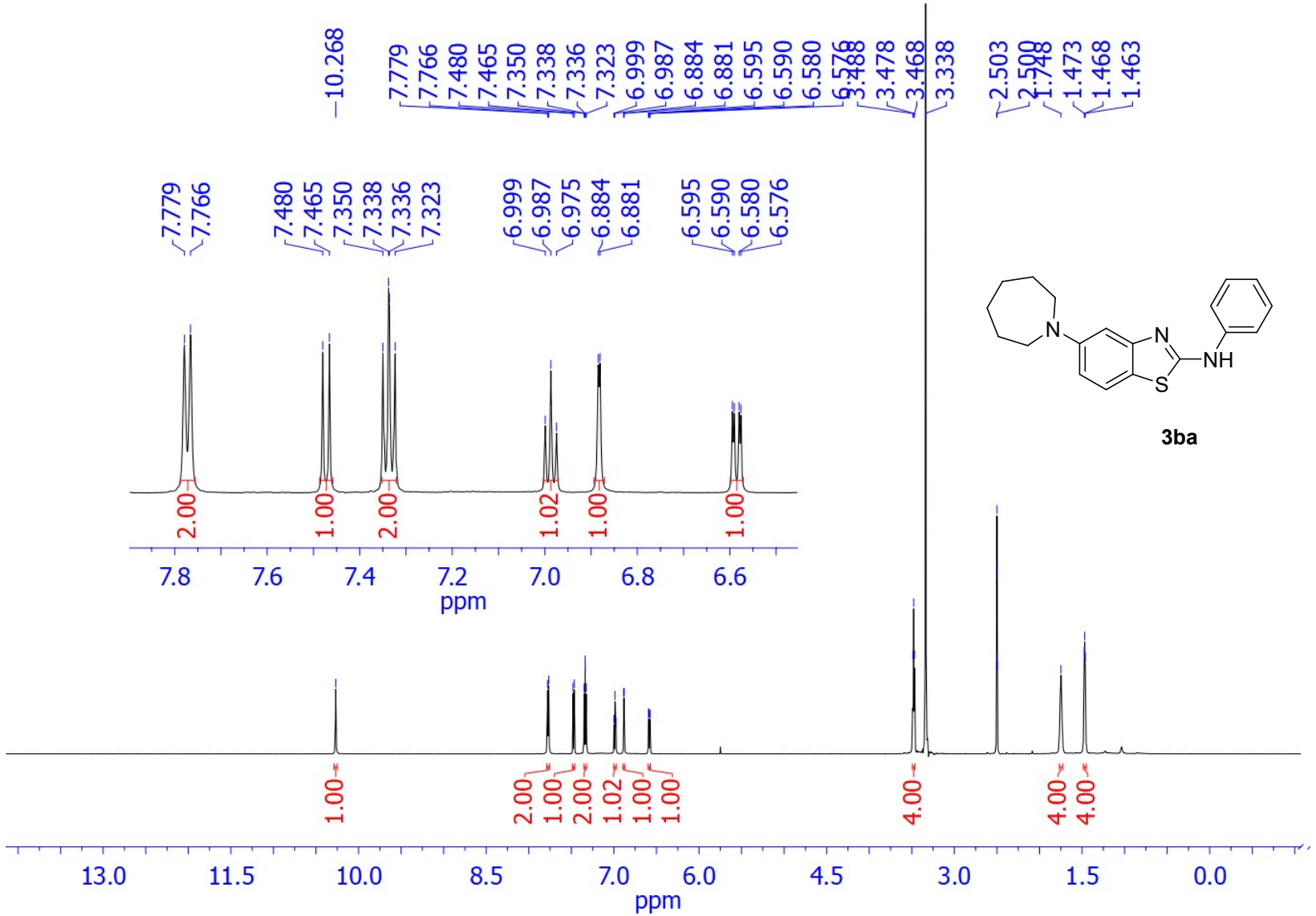
-120
-130
-140
ppm
CF3
CF3
Me2N
N
NH
S

3ah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6.881
6.595
6.590
6.580
6.576
3.478
3.468
3.338
2.503
1.748
1.473
1.468
1.463
7.779
7.766
7.480
7.465
7.350
7.338
7.336
7.323
6.999
6.987
6.975
6.884
6.881
6.595
6.590
6.580
6.576
2.00
1.00
2.00
1.02
1.00
1.00
7.8
7.6
7.4
7.2
7.0
6.8
6.6
ppm
N
N
NH
S
3ba
1.00
2.00
1.00
2.00
1.02
1.00
1.00
4.00
4.00
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10.0
8.5
7.0
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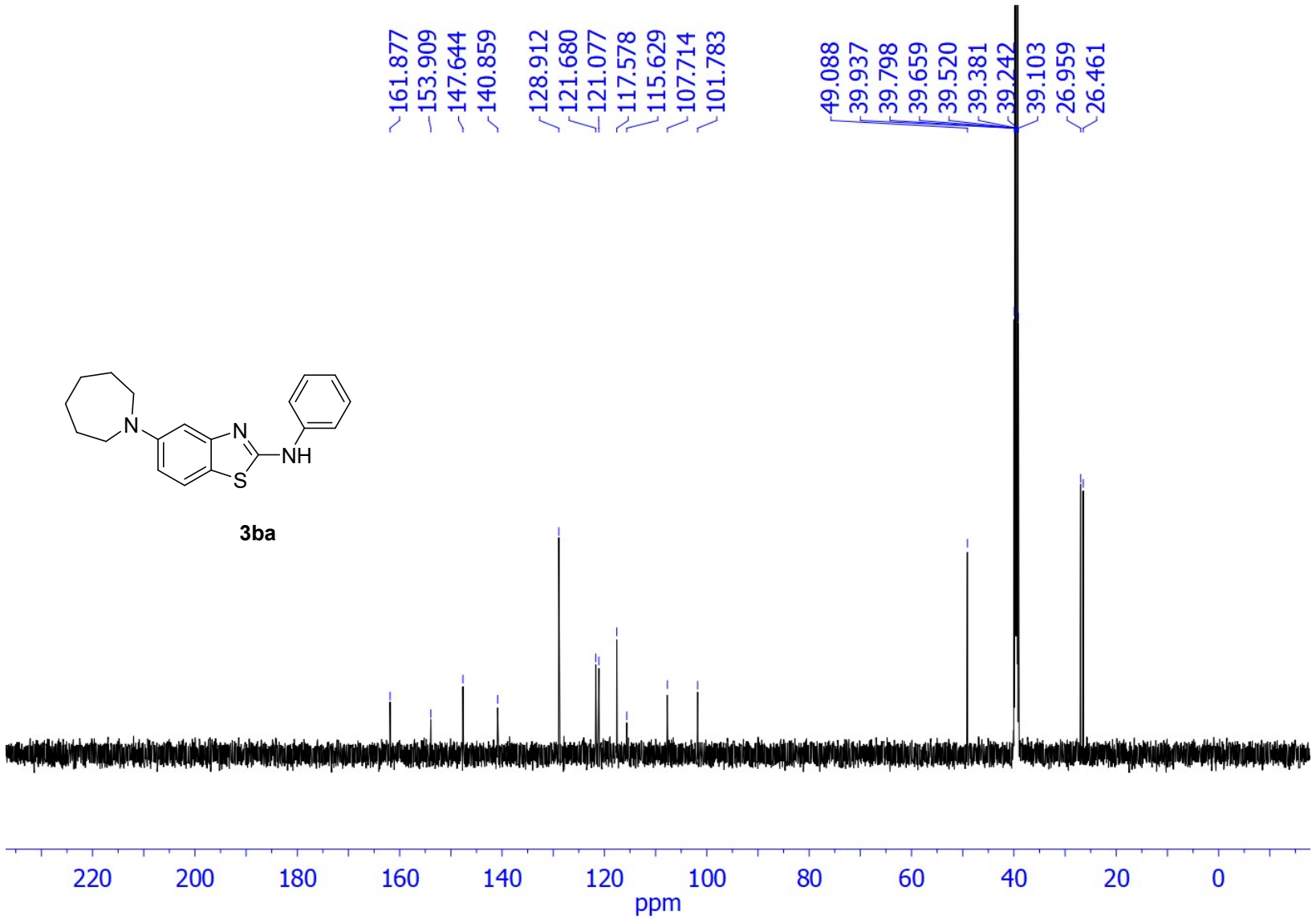
4.5
3.0
1.5
0.0
ppm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39.937
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39.659
39.520
39.381
39.242
39.103
26.959
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NH
S
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140
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100
80
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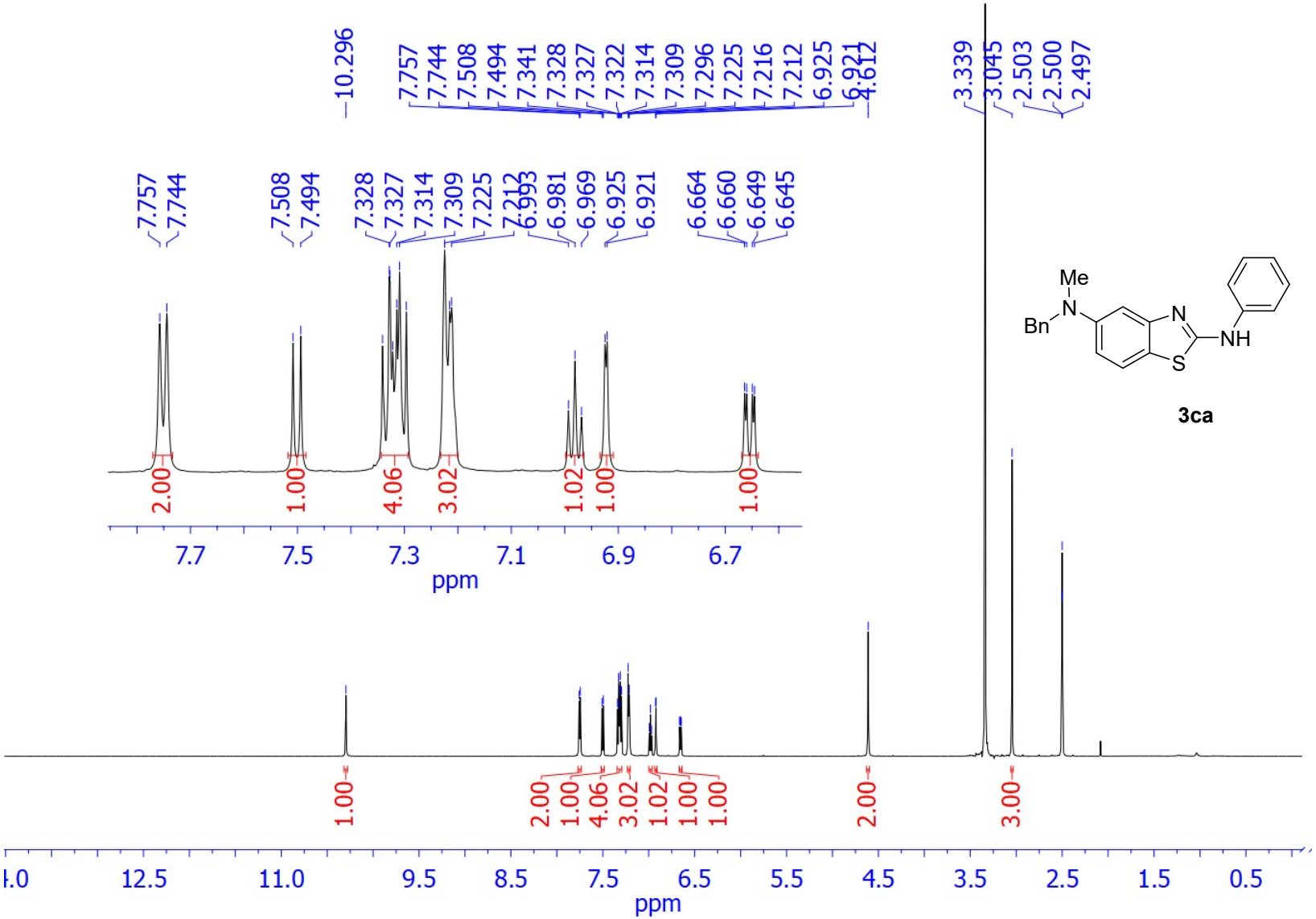
20
0
ppm

3ba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7.212
6.925
6.921
4.612
3.339
3.045
2.503
2.500
2.497
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7.744
7.508
7.494
7.328
7.327
7.314
7.309
7.225
7.212
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6.969
6.925
6.921
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6.660
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1.02
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1.00
7.7
7.5
7.3
7.1
6.9
6.7
ppm
1.00
2.00
1.00
4.06
3.02
1.02
1.00
1.00
2.00
3.00
12.5
11.0
9.5
8.5
7.5
6.5
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3.5
2.5
1.5
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ppm
Me
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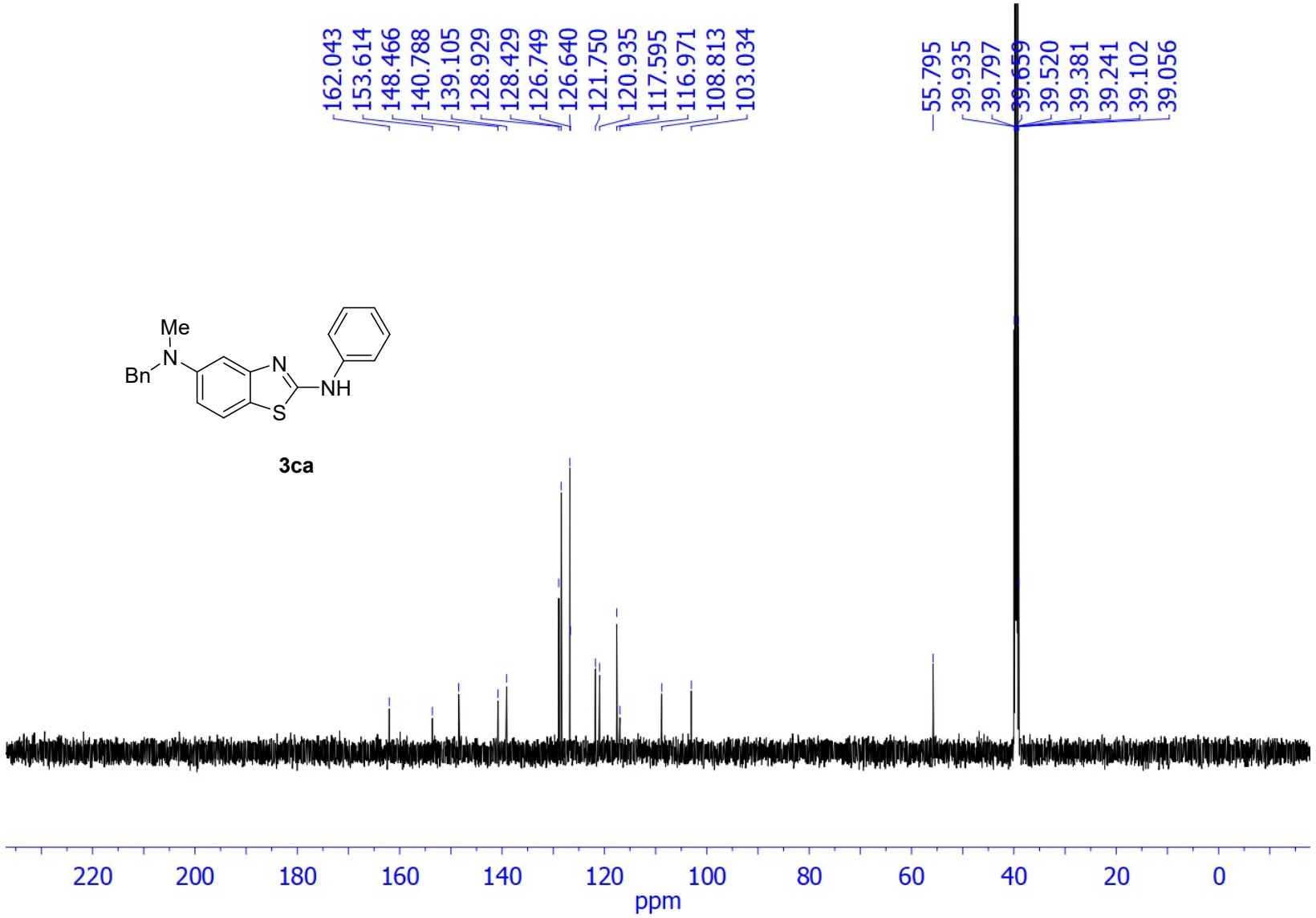
Bn
N
NH
S
3ca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39.520
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39.241
39.102
39.056
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S
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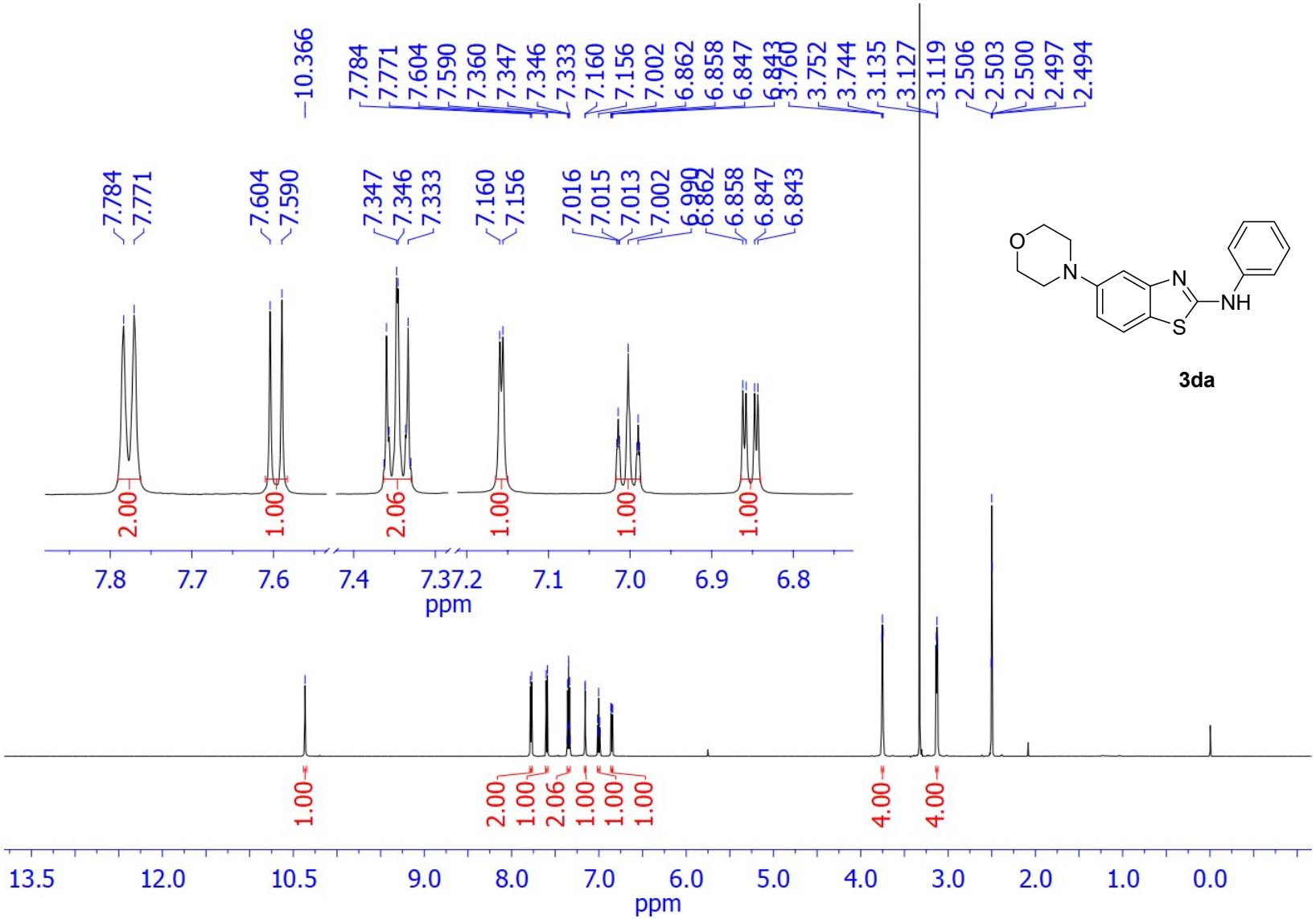
ppm

3ca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7.346
7.333
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7.156
7.002
6.862
6.858
6.847
6.843
3.760
3.752
3.744
3.135
3.127
3.119
2.506
2.503
2.500
2.497
2.494
7.784
7.771
7.604
7.590
7.347
7.346
7.333
7.160
7.156
7.016
7.015
7.013
7.002
6.862
6.858
6.847
6.843
2.00
1.00
2.06
1.00
1.00
1.00
7.8
7.7
7.6
7.4
7.37.2
7.1
7.0
6.9
6.8
ppm
O
N
N
NH
S
3da
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2.00
1.00
2.06
1.00
1.00
1.00
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13.5
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3.0
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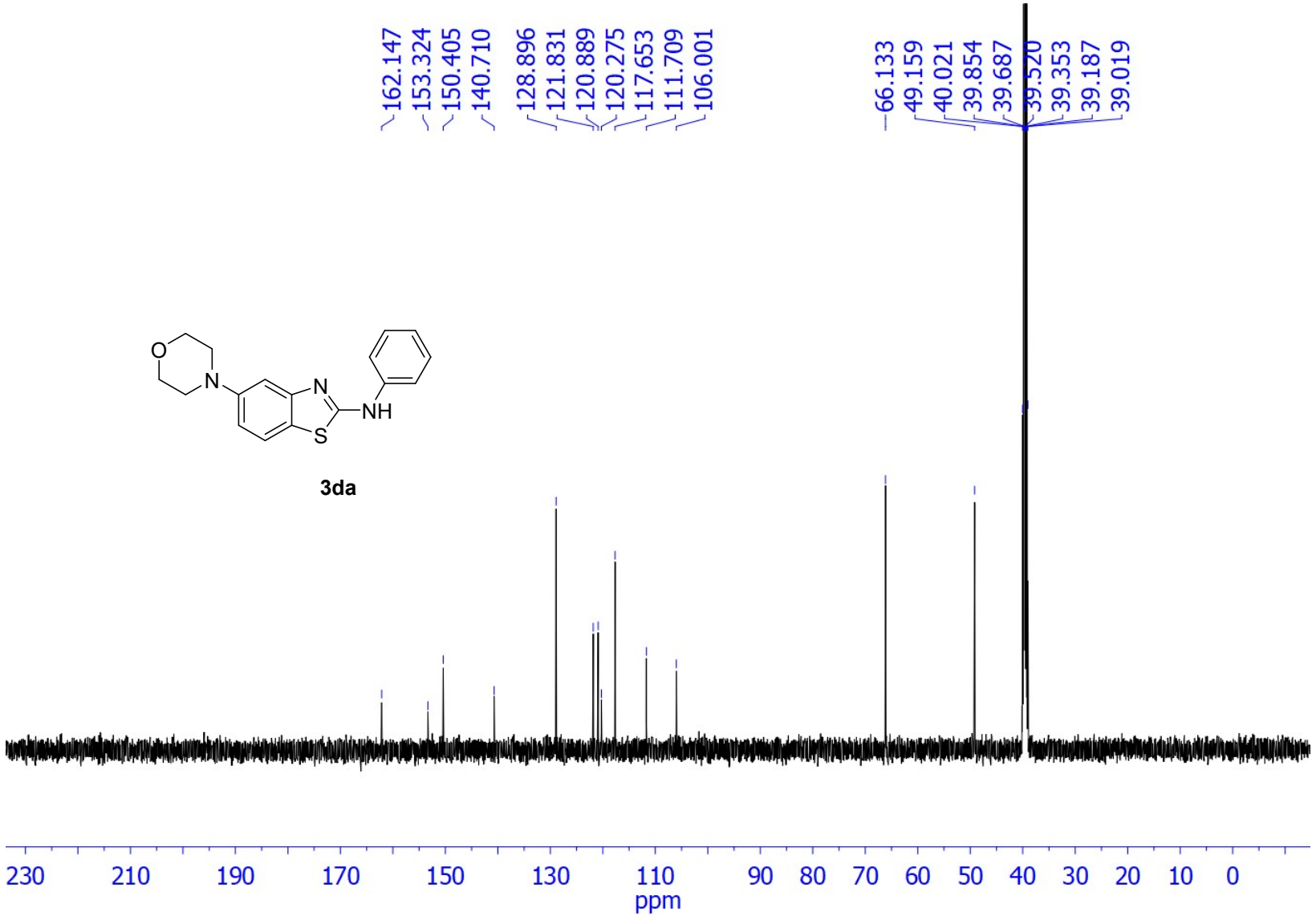
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49.159
40.021
39.854
39.687
39.520
39.353
39.187
39.019
O
N
N
NH
S
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170
150
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20
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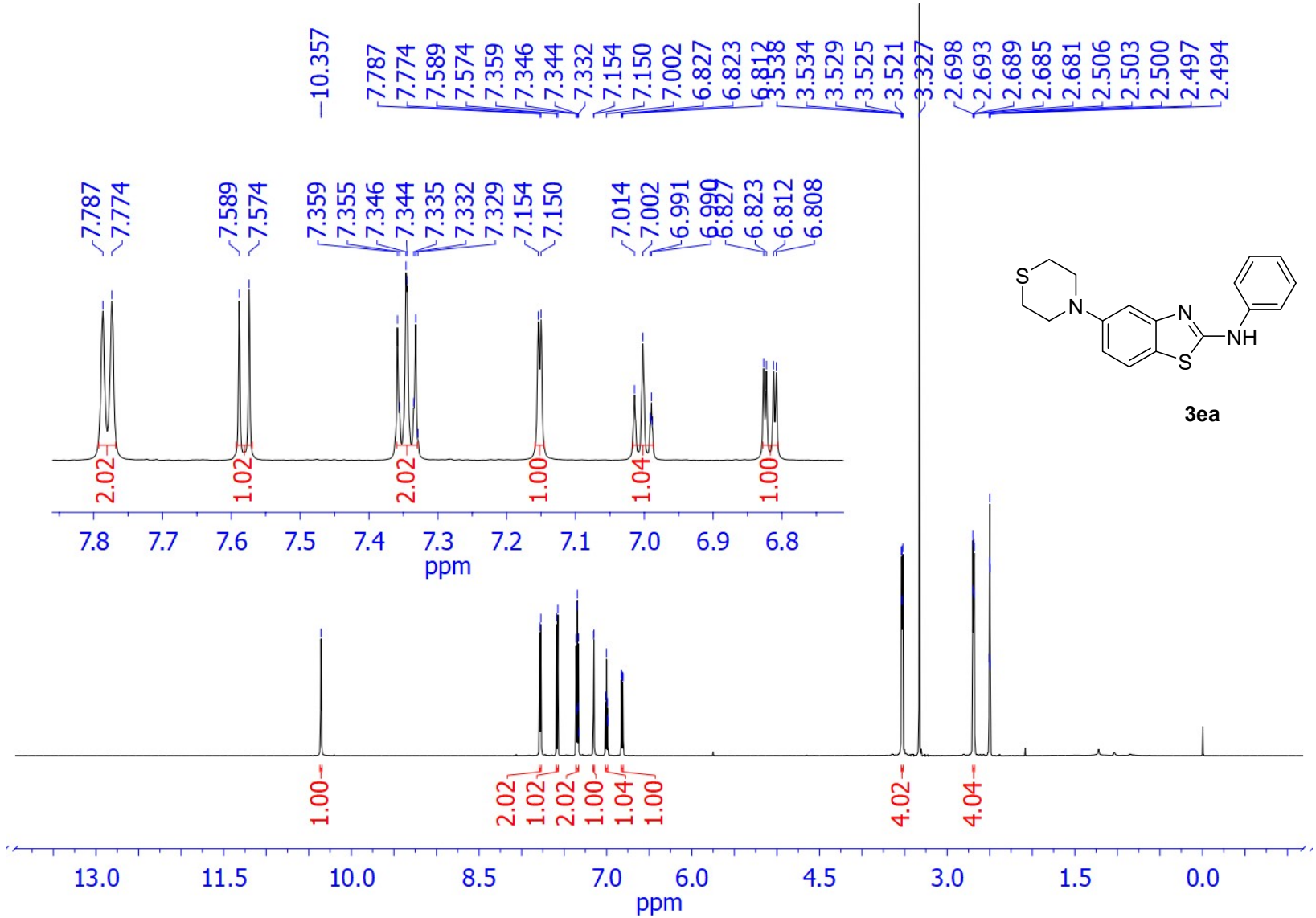
ppm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2.693
2.689
2.685
2.681
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2.503
2.500
2.497
2.494
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7.774
7.589
7.574
7.359
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7.344
7.335
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7.014
7.002
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2.02
1.00
1.04
1.00
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7.7
7.6
7.5
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7.3
7.2
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6.8
ppm
1.00
2.02
1.02
2.02
1.00
1.04
1.00
4.02
4.04
13.0
11.5
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4.5
3.0
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ppm
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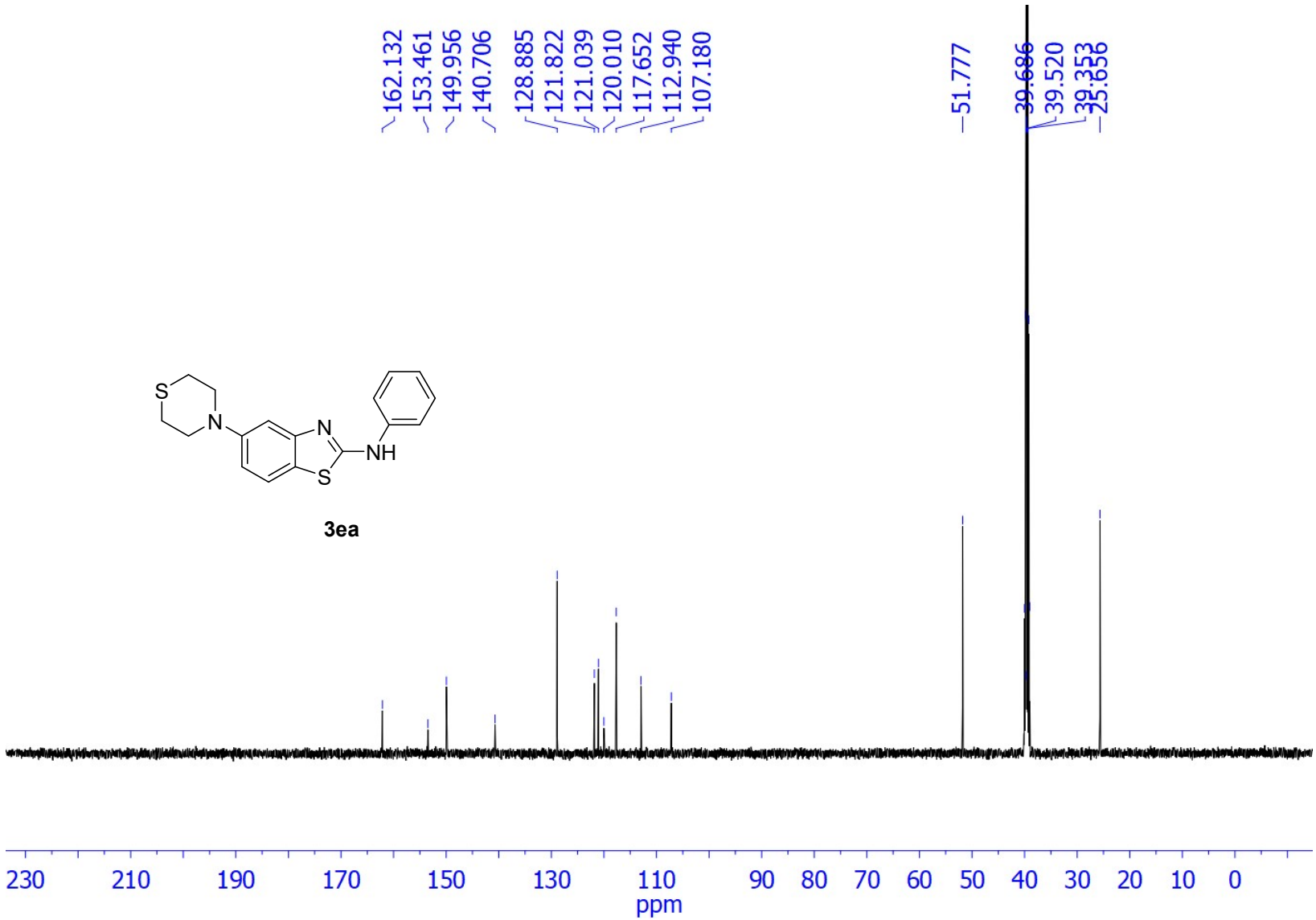
N
N
NH
S

3ea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39.520
39.353
25.656
S
N
N
NH
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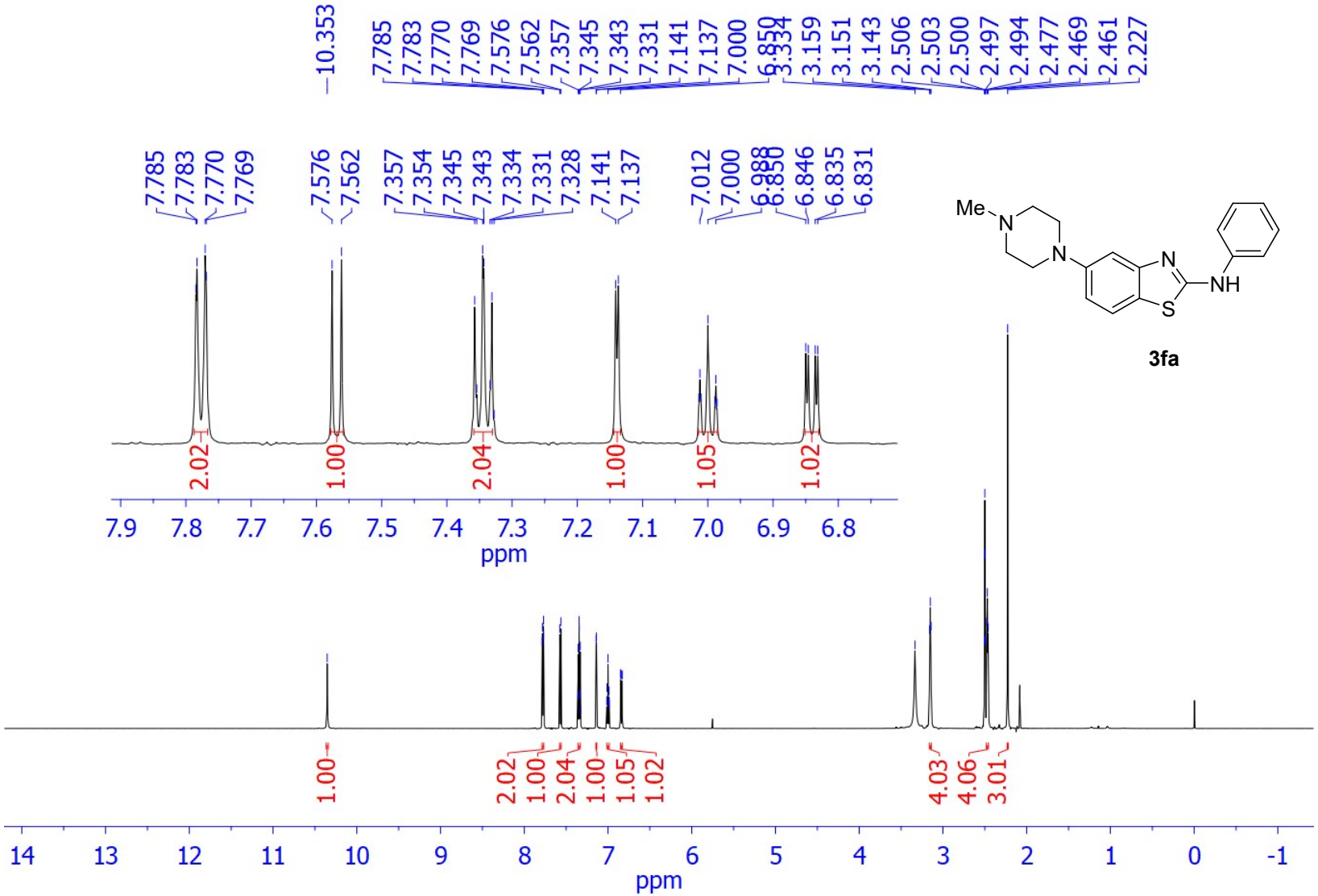
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7.343
7.331
7.141
7.137
7.000
6.850
3.334
3.159
3.151
3.143
2.506
2.503
2.500
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2.494
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2.461
2.227
7.785
7.783
7.770
7.769
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7.562
7.357
7.354
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7.343
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7.331
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6.831
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1.00
1.05
1.02
7.9
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7.7
7.6
7.5
7.4
7.3
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7.1
7.0
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6.8
ppm
Me
N
N
N
NH
S
3fa
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2.02
1.00
2.04
1.00
1.05
1.02
4.03
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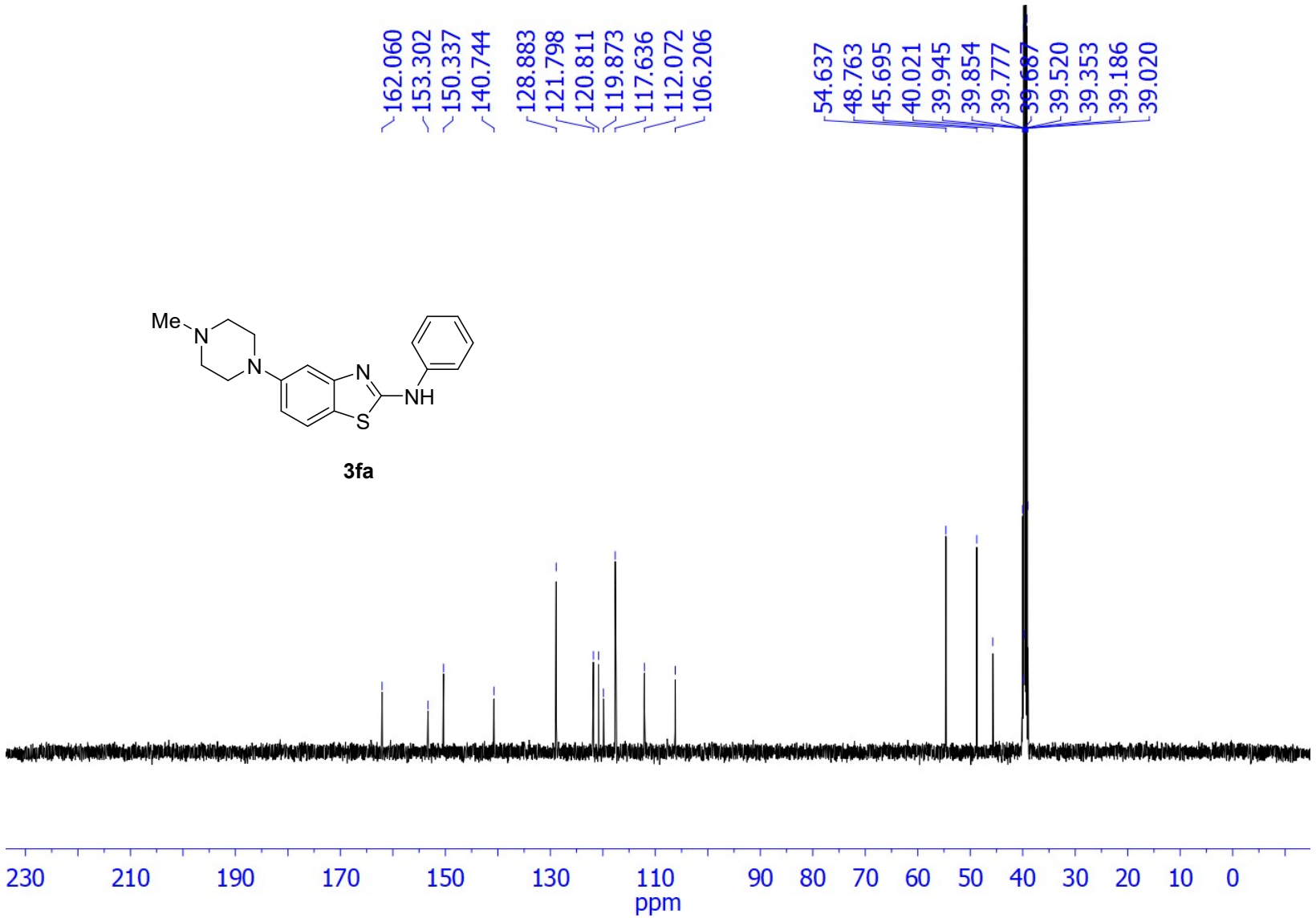
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45.695
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39.687
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39.353
39.186
39.020
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N
N
NH
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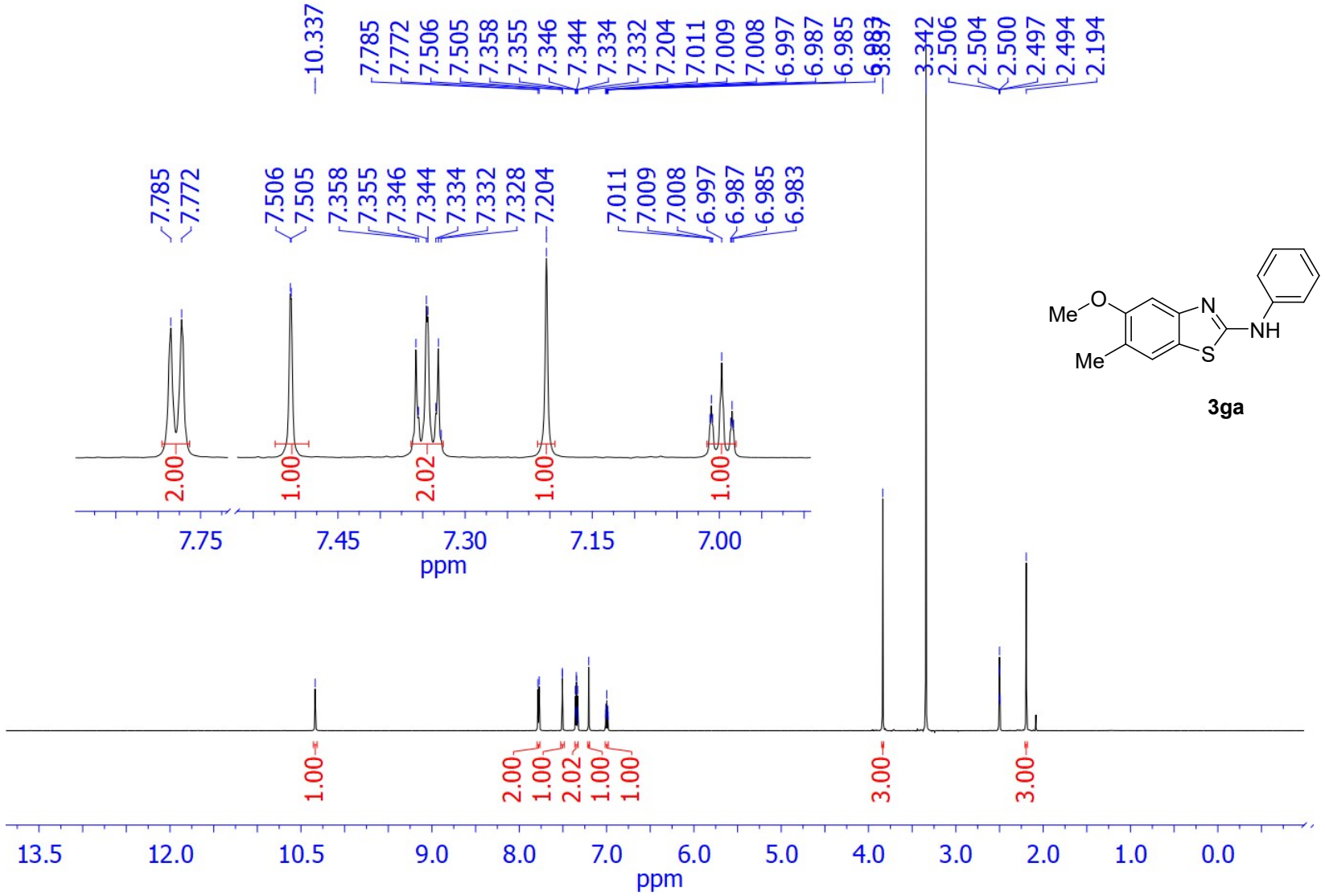
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3.00
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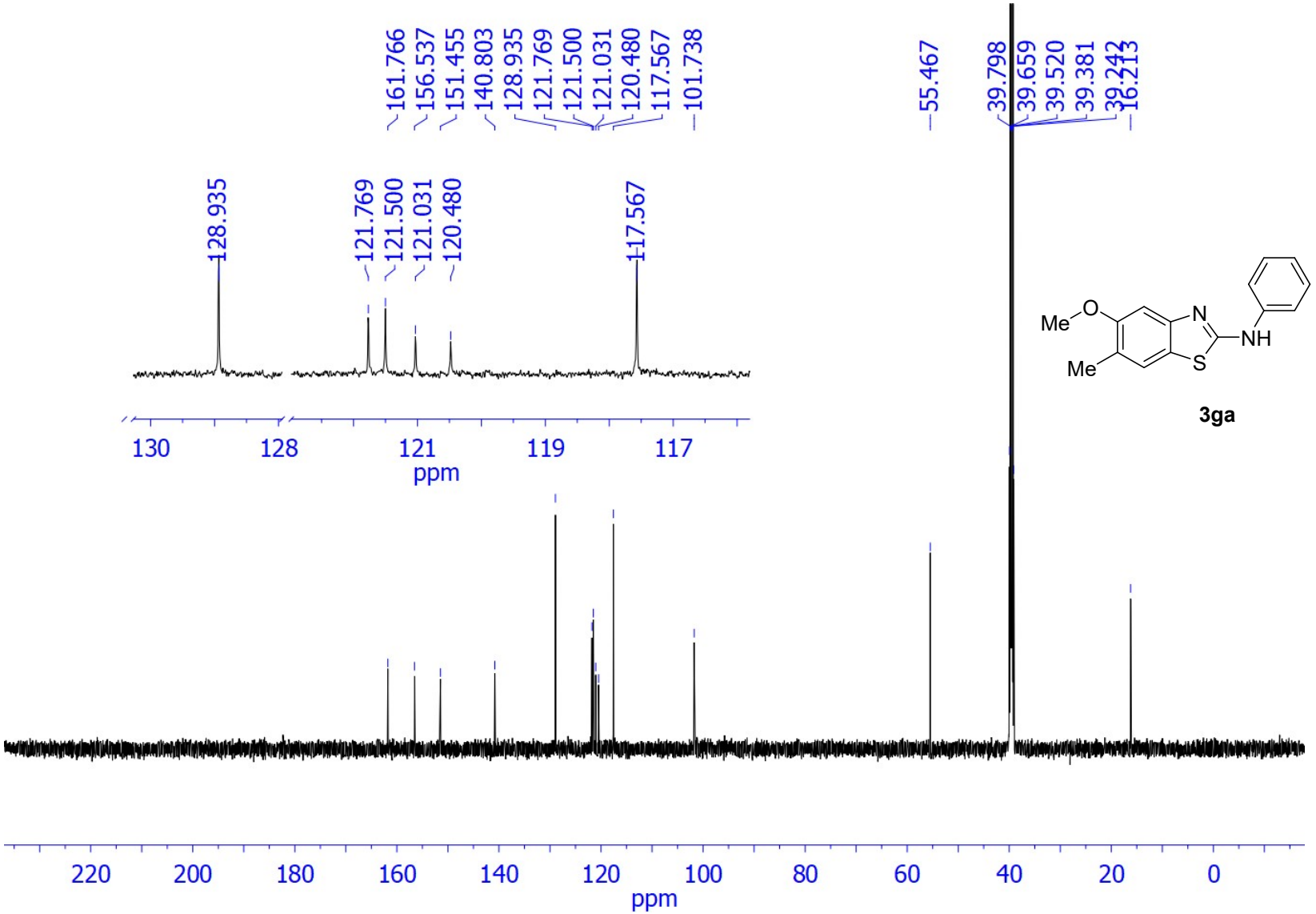
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121.769
121.500
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120.480
117.567
130
128
121
119
117
ppm
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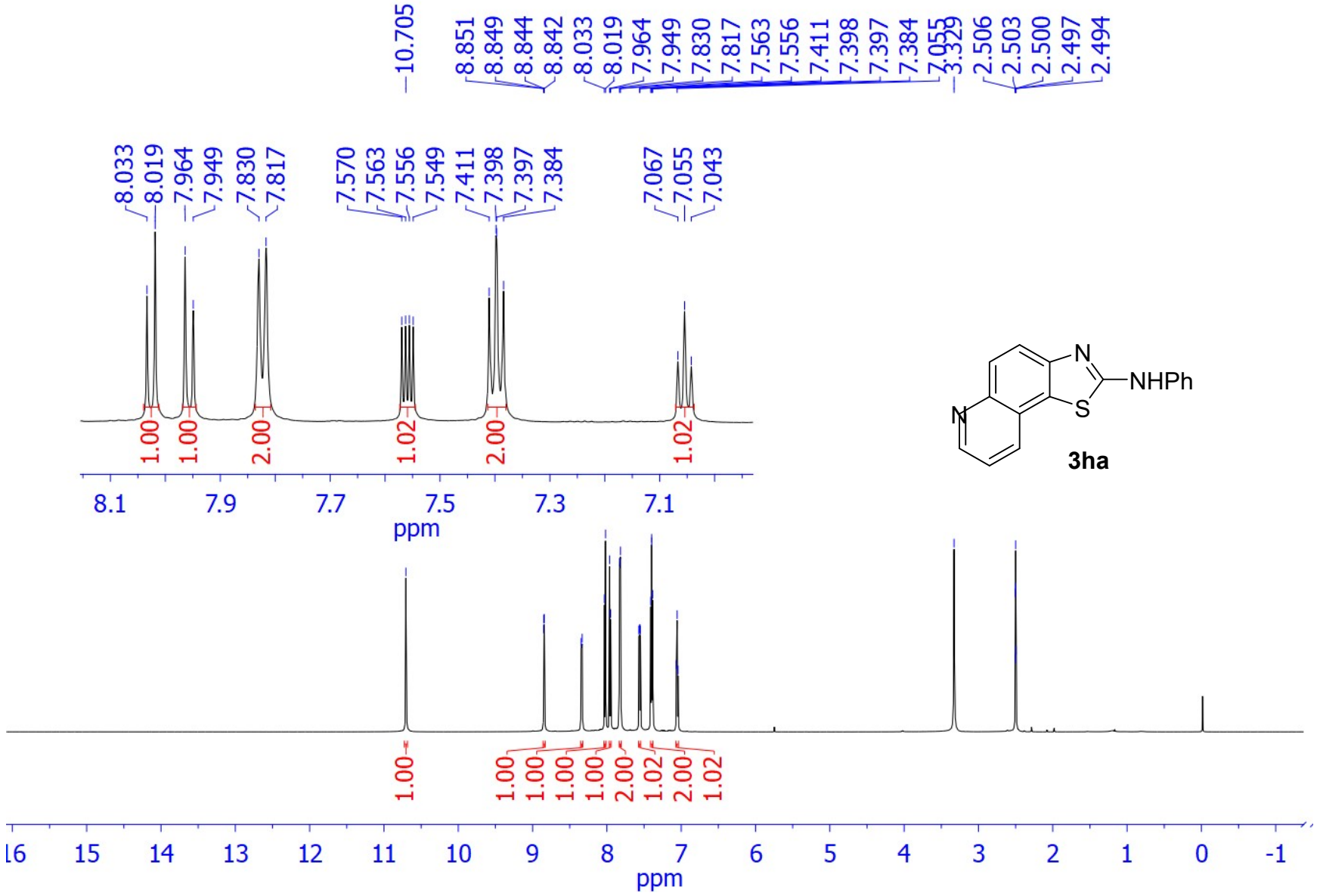
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7.055
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2.506
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2.494
8.033
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7.067
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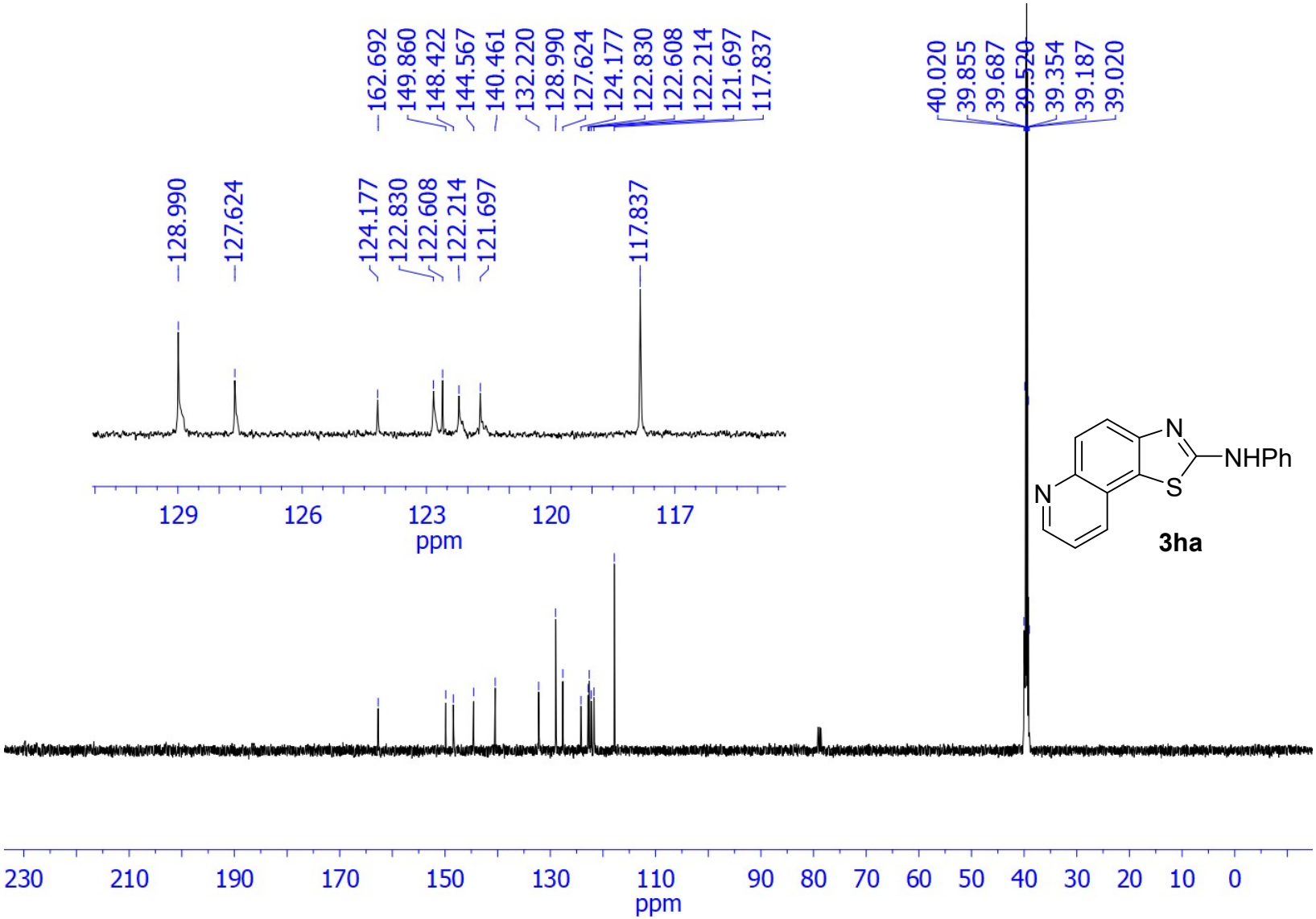
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39.020
128.990
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121.697
117.837
129
126
123
120
117
ppm
N
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S
N
3ha
230
210
190
170
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130
110
90
80
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50
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30
20
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ppm